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# THE AMERICAN MINERALOGIST

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## X-RAY IDENTIFICATION OF CHLORITE SPECIES\*

G. W. BRINDLEY AND F. H. GILLERY, *Department of Ceramic Technology,  
The Pennsylvania State University, University Park, Pa.*

### ABSTRACT

An x-ray method is developed for identifying chlorite species in fine-grained materials. A survey of the crystal chemistry of chlorites shows that in orthochlorites the principal compositional variables are the Al ions in tetrahedral coordination and the  $\text{Fe}^{2+}$  ions in octahedral coordination. These can be estimated from the basal (001) spacing and the structure factors of the lower order reflections. The problems which arise with lepto-chlorites and the applicability of the method to chlorites containing dioctahedral layers are discussed. Four examples are given comparing results obtained by the x-ray method with the results of chemical analysis.

Isomorphous substitution is exceedingly varied in the chlorite group of minerals and various schemes have been suggested for subdivision into species according to chemical composition, notably by Tschermak (1890, 1891), Orcel (1927), Orcel et al. (1950) and Hey (1954). The relations of optical properties to chemical composition have been considered especially by Winchell (1926, 1936) and also by Hey (1954), so that in the absence of chemical knowledge, species can be determined from optical data with a considerable degree of certainty. The development of an x-ray method for species identification is of interest chiefly in relation to materials which are unsuitable for chemical and/or optical study, such as clays, shales and fine-grained materials generally. Eventually, when the x-ray method has been more widely tested, it may have more general applicability for reasons of speed and convenience.

### THE NATURE OF THE PROBLEM

Since all members of the chlorite family have the same type of structure (Fig. 1), the identification of particular species must proceed from a consideration of the finer details of the x-ray diagram, namely the precise lattice dimensions and the intensities of reflections (or structure factors). Hitherto, x-ray studies had been concerned only with the lattice dimen-

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sions (Engelhardt (1942), Gruner (1944), Bannister and Whittard (1945), Brindley and MacEwan (1953), Hey (1954)) and not at all with reflected  $x$ -ray intensities. With sufficient structure factor data, the problem could be fully solved by standard methods of structure analysis, but the restrictions imposed by the use of the powder method and impure materials make it necessary to consider how much information can be extracted from the available reflections.

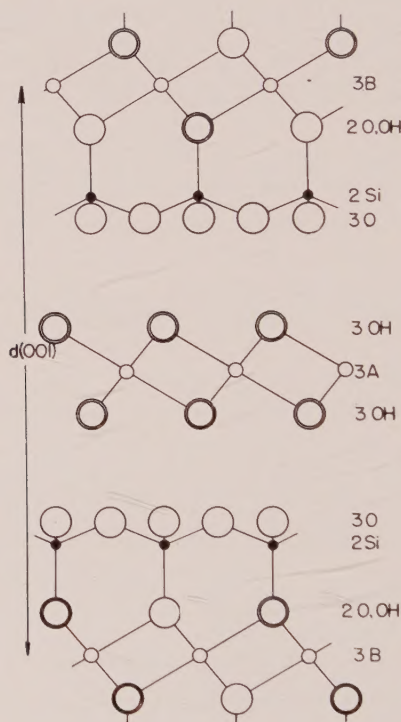


FIG. 1. The chlorite structure projected on 010. In this projection, O and (OH) ions overlap and their distribution in the  $b$  direction cannot be shown.

In the following sections we consider the principal compositional parameters of the chlorites and the application of lattice spacing and structure factor measurements to their determination. Finally a number of examples are given comparing results obtained by the  $x$ -ray method with those obtained by chemical analysis.

#### STRUCTURAL CHEMISTRY OF THE CHLORITES

The chlorite field is customarily divided into two broad subdivisions, the *orthochlorites* with compositions ranging between  $(\text{Mg}, \text{Fe}^{2+})_4\text{Al}_2(\text{Si}_2-$

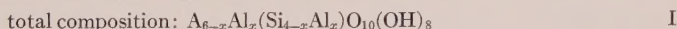
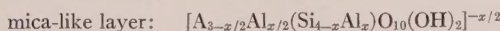
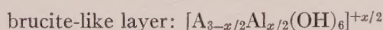


$\text{Al}_2\text{O}_{10}(\text{OH})_8$  and  $(\text{Mg}, \text{Fe}^{2+})_6\text{Si}_4\text{O}_{10}(\text{OH})_8$  with minor amounts of other elements, and the *leptochlorites* which do not fit into this scheme and are generally richer in trivalent ions, notably  $\text{Fe}^{3+}$ . In many cases the latter appear to have originated as ferrous orthochlorites and to have been subsequently oxidized (see discussion by Hey, 1954).

The chlorite structure, see Fig. 1, consists of an alternating series of brucite-like hydroxide layers, of general composition  $\text{A}_3(\text{OH})_6$ , and mica-like layers, of general composition  $\text{B}_3(\text{Si}, \text{Al})_4\text{O}_{10}(\text{OH})_2$ . A and B stand for a variety of ions in octahedral positions which, in the orthochlorites, are mainly divalent. If there are  $(\text{Si}_{4-x}\text{Al}_x)$  atoms in tetrahedral positions, the layer acquires a negative charge  $-x$  which is balanced by a corresponding number of trivalent ions in the A and B positions. On a priori grounds it is not evident whether the distribution of cations in the A and B positions is the same, and whether the charge is balanced wholly in the mica-like layer or in both layers.

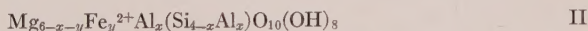
### Case I. Orthochlorites

The simplest situation arises when the A and B sites are occupied by the same distribution of cations, and Al is the only trivalent ion. The compositions and charges of the layers are then as follows:



### Case II. Orthochlorites containing Mg and $\text{Fe}^{2+}$ ions

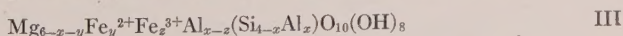
If, in formula I,  $\text{A}_{6-x}$  contains  $y$   $\text{Fe}^{2+}$  ions and the remaining ions are Mg, the formula becomes



Chlorites satisfying these conditions require the determination of two parameters,  $x$  and  $y$ .

### Case III. Orthochlorites containing $\text{Fe}^{3+}$ ions

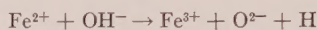
In orthochlorites,  $\text{Fe}^{3+}$  is usually a minor constituent and probably occurs wholly in octahedral positions. In the light of recent work by Osthaus (1953) on the crystal chemistry of  $\text{Fe}^{3+}$  ions in montmorillonites, the possibility cannot be entirely dismissed that  $\text{Fe}^{3+}$  ions may occupy tetrahedral positions, but if the usual assumption is made that they occupy octahedral positions, then the formula can be written:



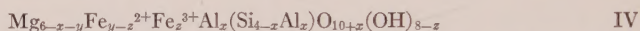
Three variables are required to specify such a chlorite.

*Case IV. Leptochlorites*

If these are oxidized chlorites (Hey, 1954) formed by a process of the type



where H leaves the lattice as  $\text{H}_2\text{O}$  by aerial oxidation, then the  $\text{Fe}^{3+}$  ions are not replacing Al ions in octahedral coordination. The leptochlorite formula can then be written

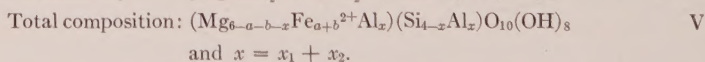
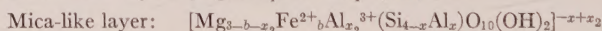
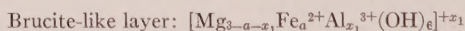


where  $z$  is the number of ferrous ions which have oxidized to ferric ions. The formula represents a partially oxidized orthochlorite of type II. When the original mineral already contains some ferric ions, as in formula III, a more complex leptochlorite formula is required.

*Case V. Chlorites with unequal distributions of octahedral cations.*

A possibility not usually considered, mainly because it is not amenable to bulk chemical analysis, is that the octahedral ions are not distributed equally between the *A* and *B* positions. It is conceivable, for example, that the replacement of divalent by trivalent cations required for electrical neutrality may occur wholly or largely within the mica-like layer. Such a structure would have little or no ionic binding force between layers and would be expected to resemble talc as regards mechanical properties.

Another possibility is that a chlorite may be formed from a mica by replacing the interlayer cations by a hydroxide layer carrying the same overall ionic charge. The ions occupying the *A* and *B* sites may then be of different compositions. If  $\text{Fe}^{3+}$  ions are absent, the compositions and charges of the component layers can be expressed as follows:



Four parameters,  $a$ ,  $b$ ,  $x_1$  and  $x_2$ , are required to describe this arrangement. Bulk chemical analysis cannot differentiate case IV from case II; it will determine  $(a+b)=y$  and  $(x_1+x_2)=x$  but it provides no means of splitting  $x$  and  $y$  into components for the two layers. In so far as  $x$ -ray analysis can solve this problem, it will give information not obtainable by ordinary chemical procedures.

*Case VI. Chlorites with dioctahedral layers*

A further possibility is that one or both of the octahedral layers in a chlorite structure may be dioctahedral rather than trioctahedral.



Possible combinations are as follows:

Brucite-like layer:  $[\text{Mg}_{3-x}\text{Al}_x(\text{OH})_6]^{+x}$   
(trioctahedral)

Mica-like layer:  $[\text{Al}_2(\text{Si}_{4-x}\text{Al}_x)\text{O}_{10}(\text{OH})_2]^{-x}$   
(dioctahedral)

Total composition:  $(\text{Mg}_{3-x}\text{Al}_{2+x})(\text{Si}_{4-x}\text{Al}_x)\text{O}_{10}(\text{OH})_8$  VIa

Still another possibility is the double dioctahedral case:

Gibbsite-like layer:  $[\text{Al}_{2+x/3}(\text{OH})_6]^{+x}$   
(approx. dioctahedral)

Mica-like layer:  $[\text{Al}_2(\text{Si}_{4-x}\text{Al}_x)\text{O}_{10}(\text{OH})_2]^{-x}$   
(dioctahedral)

Total composition:  $\text{Al}_{4+x/3}(\text{Si}_{4-x}\text{Al}_x)\text{O}_{10}(\text{OH})$  VIb

Chlorites of these compositions have not yet been discovered, but their possible existence in fine-grained sediments cannot be ignored, and it is worth considering how they might be recognized from x-ray data.

#### X-RAY EVALUATION OF CHLORITE SPECIES

The quantities experimentally determinable are the lattice parameters,  $d(001)$  and  $b$ , and the reflected intensities (or structure factors). Since chlorites have a well defined layer structure parallel to (001), the basal 00 $l$  reflections will provide the clearest information on the ionic distribution.

##### *The b parameter*

This parameter is best determined from the easily recognized 060 reflection, with  $d$  about 1.53–1.54 Å for all trioctahedral layer silicates. The 040 reflection is usually weak and not easily seen in a powder diagram and the 020 reflection is often obscured by the 003 reflection. The main usefulness of the  $b$  parameter is that it provides a clear distinction between di- and trioctahedral structures.

Within the trioctahedral group  $b$  is not well suited to the recognition of particular species. Engelhardt (1942) and Hey (1954) correlated  $b$  with the number of Fe atoms in the chlorite structure, but Brindley and MacEwan (1953, pp. 38–41) have shown that it must be related to all the lattice substitutions. Since  $\text{AlO}_4$  tetrahedra are larger than  $\text{SiO}_4$  tetrahedra, increasing substitution of Si by Al tends to expand the tetrahedral layers. If Mg ions are regarded as the normal octahedral cations, then replacement of  $\text{Mg}^{2+}$  (ionic radius, 0.78 Å) by  $\text{Al}^{3+}$  and  $\text{Fe}^{3+}$  (ionic radii 0.57 and 0.67 Å respectively) tends to shrink the lattice, and replacement by  $\text{Fe}^{2+}$  ions (ionic radius, 0.83 Å) to expand it. Since the tetrahedral and octahedral layers are parallel to one another, the actual  $b$

parameter will be the result of a compromise between the two kinds of layers and therefore a function of all the substitutions within the lattice. In so far as Al ions occupy tetrahedral and octahedral positions equally, their overall contribution to an expansion or contraction of the layers may be small and the resultant  $b$  value may then be determined principally by the  $\text{Fe}^{2+}$  substitutions. However, we consider that  $b$  is not a useful parameter to use for species determination, and that the Fe content can be more reliably determined from structure factor determinations.

*The basal spacing,  $d(001)$*

This spacing depends on the compositions of the layers and the bonding forces between them. Gruner (1944) and Bannister and Whittard

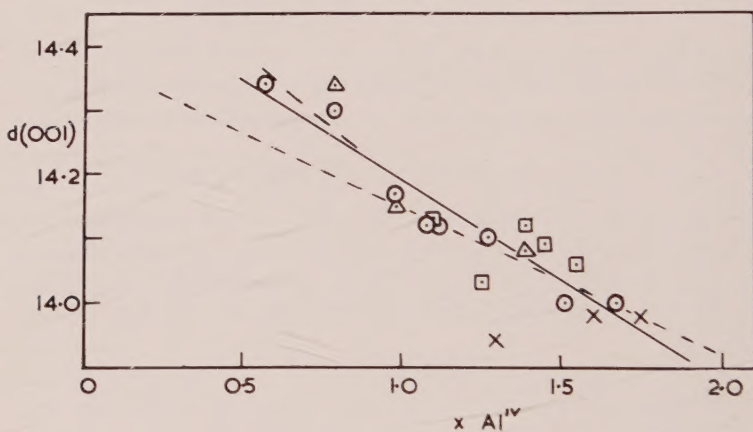


FIG. 2. Basal spacing,  $d(001)$ , of chlorites plotted against  $x$ , the number of Al atoms replacing Si atoms in the formula  $(\text{Si}, \text{Al})_4$ . Experimental observations are as follows:  $\circ$  Bannister and Whittard (1945);  $\times$  Engelhardt (1942);  $\square$  McMurchy (1934);  $\triangle$  Brindley and Ali (See Brindley, 1951). Dashed line shows a relation given by Hey (1954).

(1945) correlated the basal spacing with the replacement of Si by Al, and attributed the variation of  $d(001)$  principally to the ionic bonding forces. In orthochlorites satisfying formulae II and III, the ionic bonding forces depend mainly on the layer charges,  $\pm x/2$ , where  $x$  = no. of Al ions in tetrahedral positions. Substitutions within the layer determine their dimensions in the manner already indicated, but if these dimensions remain roughly constant, then  $d(001)$  can be correlated with  $x$  as a first approximation.

In Fig. 2, observed values of  $d(001)$  are plotted against  $x$  for a number of analyzed chlorites and the results lie on or near the straight line

$$d(001) = 14.50 - 0.31x.$$



Hey (1954) has given a similar relation with constants 14.38 and  $-0.23$  in the present notation, which is also shown in Fig. 2.

We have examined the scatter of the points in Fig. 2 in relation to the  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  contents of the minerals and, in agreement with Hey, find a tendency for Fe-rich chlorites to give low spacings but the observations are too scattered to justify at this stage an attempt to express quantitatively how the mean line of Fig. 2 varies with Fe content.

#### *The intensities of the basal reflections*

No quantitative discussion of the basal intensities has previously been given although it is well-known that the (001), (003) and (005) reflections are relatively weak from iron-rich chlorites. This follows directly from the fact that the octahedral layers associated with the A and B cations are separated by a distance of  $d(001)/2$ , so that the only contributions to the odd orders come from the Si-O hexagonal networks. The octahedral layers make additive contributions to the even orders which are relatively very strong for the iron-rich chlorites. For the same reasons it is seen directly that an unequal distribution of cations between the A and B sites leaves the even order basal reflections unaffected, but has a pronounced effect on the odd orders because the contributions from the octahedral layers no longer cancel out exactly.

The nearly equal scattering factors of Si, Al and Mg, and of Fe, Cr and Mn preclude any distinction between atoms in these groups on the basis of intensity measurements. It should be possible, however, to locate and estimate Fe (and equivalent) atoms in relation to Mg (and equivalent) atoms. Structure factors have been calculated for a range of chlorite compositions using atomic scattering factors given by Bragg and West (1928), with allowance for  $x$ -ray dispersion in the case of the Fe atoms. Dispersion lowers the effective atomic scattering factor of Fe by amounts 1.5, 4.4 and 3.0 for Cu, Co and Fe  $K\alpha$  radiations respectively. We have used a dispersion correction of 3.0 units which is correct for Fe  $K\alpha$  radiation (this is most commonly employed for iron-rich materials) and will not be seriously in error for the other two radiations. The  $c$ -parameters for the chlorite structure have been taken from a fourier synthesis for penninite (Brindley and Robinson, 1951, p. 180); the variations of these parameters for other chlorites will have a negligible influence on the lower order structure factors.

#### RESULTS OF STRUCTURE FACTOR CALCULATIONS

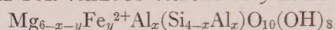
The calculated structure factors,  $F$ , tabulated in Tables 1, 2 and 3, correspond to the formulae I-VI previously given. These formulae represent the content of half of a unit cell comprising one chlorite layer. The

signs of  $F$  correspond to an origin placed at the center of the mica-like layer.

(a) *Orthochlorites without  $\text{Fe}^{3+}$  ions*

Table 1 lists the calculated  $F$  values for orthochlorites satisfying formula II and having equal cation distributions in the  $A$  and  $B$  positions.

TABLE 1(A). STRUCTURE FACTORS,  $F$ , FOR ORTHOCHLORITE REFLECTIONS  
CALCULATED FOR VARIOUS VALUES OF  $y$  IN THE FORMULA:



ASSUMING A SYMMETRICAL DISTRIBUTION OF THE OCTAHEDRAL CATIONS.

THE VALUES OF  $F$  CORRESPOND TO ONE FORMULA UNIT

$y \backslash 00l$	001	002	003	004	005	006	007	008
0	22	41	-55	80	55	-18	-40	-11
1	22	51	-55	88	55	-2	-40	-6
2	22	61	-55	96	55	-5	-40	-1
3	22	71	-55	104	55	1	-40	4
4	22	80	-55	111	55	7	-40	9
5	22	90	-55	119	55	13	-40	14
6	22	100	-55	127	55	19	-40	19

( $F$  values for odd orders are independent of  $y$ .)

TABLE 1(B). STRUCTURE FACTOR RATIOS

$y$	$F(002)/F(001)$	$F(002)/F(003)$	$F(004)/F(002)$	$F(004)/F(003)$ $F(004)/F(005)$
0	1.86	0.75	1.95	1.45
1	2.32	0.93	1.73	1.60
2	2.77	1.11	1.57	1.75
3	3.22	1.29	1.46	1.89
4	3.64	1.45	1.39	2.02
5	4.09	1.64	1.32	2.16
6	4.55	1.82	1.27	2.31

$F$  is independent of  $y$  (the number of  $\text{Fe}^{2+}$  ions) for the odd order reflections, but increases with  $y$  for the even orders. The parameter  $y$  is determined from  $|F|$  values measured in arbitrary units by comparing observed and calculated values of the ratios  $F(002)/F(001)$ ,  $F(002)/F(003)$ ,  $F(004)/F(002)$ ,  $F(004)/F(003)$  and  $F(004)/F(005)$ . Although higher orders are included in Table 1, it is seldom they will be available in actual



analyses due to the presence of other reflections, except when pure chlorites are examined.

The accuracy with which  $y$  can be determined by this procedure rests on several considerations. On the experimental side, the reflected intensities must be accurately measured and correctly converted to  $|F|$  values. This requires careful attention to points of detail to ensure that the experimental conditions presupposed by a theoretical formula are adequately satisfied. On the theoretical side, the calculated values depend on the accuracy of the scattering factors and the state of ionization of the atoms in the structure.

(b) *Orthochlorites containing  $Fe^{3+}$  ions*

Formula III represents the case where the  $Fe^{3+}$  ions are wholly in octahedral positions, and the octahedral cations are equally distributed. X-ray intensities do not distinguish between  $Fe^{2+}$  and  $Fe^{3+}$ , and the use of Table 1 therefore gives  $(y+z)$ . The parameter  $x$  will be obtained from Fig. 1 and this gives the amount of Al in tetrahedral positions. If, however, as in case (a), we take  $x$  instead of  $(x-z)$  for the octahedral Al, then octahedral Al is over-estimated and the Mg will be under-estimated.

At present there is no obvious way by which the  $z$   $Fe^{3+}$  ions in formula III can be determined by x-rays. However, it appears to be the case that  $Fe^{3+}$  is usually a small component in orthochlorites so that the errors in Al and Mg may be relatively unimportant.

(c) *Leptochlorites containing  $Fe^{3+}$  ions*

In so far as leptochlorites are oxidized forms of orthochlorites, they are represented by formula IV. The  $Fe^{3+}$  ions, being derived from  $Fe^{2+}$  ions, are not replacing  $Al^{3+}$  ions in octahedral sites as in case (b). The parameter  $x$  is determined from  $d(001)$  measurements, and the total iron,  $Fe_{y-z}^{2+}Fe_z^{3+}$ , will be given by use of Table 1. We still cannot determine the parameter  $z$ , but this will not introduce errors into the Mg and octahedral Al determinations, as in case (b). In effect, the result yielded by the x-ray method will be the original, unoxidized orthochlorite.

If the leptochlorite is derived from an orthochlorite containing  $Fe^{3+}$  ions in its original composition, then the same errors will arise as we have discussed in case (b).

(d) *Chlorites with unequal distributions of octahedral cations*

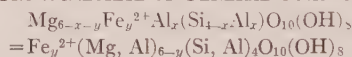
X-ray intensities will distinguish only between Fe (and equivalent) atoms and Mg (and equivalent) atoms. If the octahedral Al ions are unequally distributed in a purely magnesian chlorite, so that the  $Al^{VI}:Mg$  ratio differs in the two layers, then x-ray intensity measurements will give

no indication of this situation. Such a distribution would influence the layer charges and presumably also the  $d(001)$  spacing. In such circumstances, the use of Fig. 2 and Table 1 to determine  $x$  and  $y$  may lead to incorrect results.

On the other hand, if the unequal distribution affects the  $\text{Fe}^{2+}:\text{Mg}$  ratio, the structure factors will be sensitive indicators. Since the even order  $00l$  reflections depend on the amplitude sum from the octahedral layers and the odd order reflections on the amplitude difference, the even orders will determine the total  $\text{Fe}^{2+}$  and Mg in the octahedral positions, while the odd orders will give the asymmetry in the distribution.

Table 2 gives the results of calculations of the odd-order  $00l$  structure factors for a range of asymmetrical distributions of  $\text{Fe}^{2+}$  ions. Owing to

TABLE 2. STRUCTURE FACTORS OF ODD ORDER  $00l$  REFLECTIONS FOR VARIOUS ASYMMETRIC DISTRIBUTIONS OF  $\text{Fe}^{2+}$  IONS BETWEEN THE  $A$  AND  $B$  OCTAHEDRAL POSITION IN AN ORTHOCHLORITE OF GENERAL COMPOSITION:



Distribution of $\text{Fe}^{2+}$ ions			Structure factor, $F$			
$\text{Fe}^{2+}$ ions in mica-like layer	$\text{Fe}^{2+}$ ions in brucite-like layer	Excess Fe in mica-like layer	001	003	005	007
$y/2 + \frac{1}{2}$	$y/2 - \frac{1}{2}$	1.0	34	-45	62	-34
$y/2 + \frac{1}{4}$	$y/2 - \frac{1}{4}$	0.5	28	-50	58	-37
$y/2$	$y/2$	0	22	-55	55	-40
$y/2 - \frac{1}{4}$	$y/2 + \frac{1}{4}$	-0.5	16	-60	51	-43
$y/2 - \frac{1}{2}$	$y/2 + \frac{1}{2}$	-1.0	12	-64	48	-46

the relatively large scattering factor of the Fe atom compared with those of Mg and Al, any asymmetry in the iron distribution has a very marked effect on the structure factors. Moreover, Table 2 shows that as Fe atoms are transferred from the mica-like layer to the brucite-like layer,  $|F|$  diminishes for the 001 and 005 reflections, but increases for the 003 and 007 reflections.

#### (e) Chlorites with dioctahedral layers

Table 3 lists the calculated results for two of these structures. A double dioctahedral structure (formula VIb, and No. 2 in Table 3) gives the same odd-order scattering factors as the symmetrical trioctahedral orthochlorites, (c. f. Tables 1 and 2). The even order scattering factors, particularly those for 002 and 004, bear a different ratio to each other and to the odd orders as compared with the orthochlorite data in Table 2.



Thus  $F(004)/F(002)$  is about 3.0 as compared with ratios ranging from about 1.2 to 2.0 in Table 2, the (001) and (002) reflections are more nearly equal and (003) is relatively large as compared with the orthochlorite data.

A structure which is trioctahedral in the brucite-like layer and dioctahedral in the mica-like layer, (formula VIa and No. 1 in Table 3) is characterized by a (003) reflection which is relatively strong compared with the (001) and (002) reflections. In this respect it resembles an orthochlorite with an asymmetric distribution of octahedral cations and a preponderance of Fe in the brucite-like layers, (c.f. Tables 1 and 2).

It will probably always be difficult to decide with certainty purely from x-ray intensity data if a chlorite-type mineral has one or both octahedral layers of the dioctahedral type. When such a question arises, useful auxiliary information will be provided by the  $b$  parameter. For dioctahedral layer minerals,  $b$  is usually of the order of 8.85–9.00 Å

TABLE 3. STRUCTURE FACTORS FOR CHLORITES CONTAINING DIOCTAHEDRAL LAYERS

(1) Trioctahedral brucite layer, dioctahedral mica layer, formula VIa.

(2) Dioctahedral gibbsite layer, dioctahedral mica layer, formula VIb.

00 $l$	001	002	003	004	005	006	007	008
(1)	12	31	—65	71	46	—25	—47	—17
(2)	22	21	—55	62	55	—33	—40	—24

and for trioctahedral minerals is distinctly larger, and of the order of 9.2–9.3 Å.

#### A SUMMARY OF THE PROCEDURE FOR DETERMINING CHLORITE SPECIES FROM X-RAY DATA

The spacings and reflected intensities of the first five basal 00 $l$  reflections must be accurately measured. The 060 reflection should be recorded to confirm if the mineral is trioctahedral. Care must be taken to use a valid formula for converting reflected intensities to structure factors. Geiger counter diffractometers will be found more accurate and convenient for obtaining the required data but particular care must be exercised in deriving the structure factors, (see notes in appendix).

The basal parameter  $d(001)$  in conjunction with Figure 2 gives  $x$ , the number of tetrahedral Al ions, which is also the number of octahedral Al ions except when  $\text{Fe}^{3+}$  ions substitute for Al in octahedral sites.

We have seen that  $F(001)$ ,  $F(003)$  and  $F(005)$  vary with the asymmetry

of distribution of Fe ions. For *symmetrical* distributions, these structure factors are independent of Fe content and good agreement should be obtained between the observed values and the calculated values in Table 1. If good agreement is not obtained, and provided the experimental data are reliable, then the distribution may be *asymmetrical*; the asymmetry can be considered in the light of the data given in Table 2.

When the experimental evidence points to a symmetrical distribution, then the first five 00*l* structure factors can be used in conjunction with Table 1 to determine the  $\gamma$  parameter, i.e., the number of Fe atoms. When there is doubt about the octahedral cations being symmetrically distributed, then the ratio  $F(004)/F(002)$  gives a value for  $\gamma$  which is independent of any asymmetry.

#### EXAMPLES OF THE APPLICATION OF THE METHOD

A detailed account will be given of the application of the foregoing methods to the identification of a chlorite supplied by the Sierra Talc

TABLE 4. EXPERIMENTAL DATA FOR THE IDENTIFICATION OF A CHLORITE

00 <i>l</i>	<i>d</i> (00 <i>l</i> )	<i>d</i> (001)	<i>I</i> (integrated)	<i>F</i> (relative values)
001	14.1 (4)	14.1 (4)	318	9.7 <sub>2</sub>
002	7.08	14.16	360	20.8 <sub>4</sub>
003	4.720	14.16	259	26.8 <sub>1</sub>
004	3.541	14.16	285	38.1 <sub>5</sub>
005	2.836	14.18	77 $\frac{1}{2}$	25.30
007	2.023	14.16	—	—
00, 10	1.417	14.17	—	—
00, 11	1.288	14.17	—	—
Mean		14.16 <sub>5</sub>		

and Clay Co. with the kind assistance of Professor J. A. Pask. The material was an almost pure chlorite and more basal reflections were recorded than would usually be possible with less pure material. The identification, however, has been based essentially on the first five orders. The experimental data are given in Table 4. The mean basal spacing,  $14.16_5 \pm 0.00_5$  Å, would not be materially changed by using the first five orders only. Using Fig. 2, we obtain

$$x = 1.09 \pm 0.05$$

The uncertainty in  $x$ , 0.05, is based primarily on an estimated uncertainty in the position of the line in Fig. 2. An uncertainty of  $\pm 0.01$  Å in  $d(001)$  has a relatively small effect on  $x$ .



We consider next the values of  $|F|$  for (001), (003) and (005), and obtain the following ratios:

	$ F (001)$	$ F (003)$	$ F (005)$
Experimental ratio:	0.362	1	0.94 <sub>3</sub>
Calculated ratio, assuming symmetrical distribution of octahedral cations, (Table 1)	0.40	1	1.00
Calculated ratio, assuming an excess of Fe atoms in mica layer of $-0.2$ , (using Table 2)	0.35	1	0.94

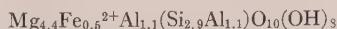
The agreement between the observed ratios and those calculated on the assumption of a symmetrical distribution of octahedral cations is quite close and it is scarcely worth while to consider an asymmetrical distribution. Nevertheless, reference to Table 2 shows that the slightly lower observed ratios are consistent with slightly less Fe in the mica layer than in the brucite layer. In fact, by transferring as little as 0.1 Fe from the mica to the brucite layer we obtain complete agreement with the observed values. This asymmetry is so small that we hesitate to claim it as a real deviation from a symmetrical distribution.

Neglecting this small asymmetry and using the full range of experimental data up to (005) in conjunction with Table 1, we obtain the following values for  $\gamma$ :

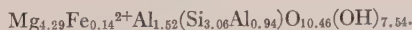
	$F(002)/F(001)$	$F(002)/F(003)$	$F(004)/F(002)$	$F(004)/F(003)$	$F(004)/F(005)$
Experimental	2.14	0.78	1.83	1.42	1.51
$\gamma$ (Table 1)	0.61	0.33	0.55	$-0.20$	0.40

Mean  $\gamma$  (excluding  $F(004)/F(003)$ ) =  $0.47 \pm 0.11$  or  $0.5 \pm 0.1$

Using formula II, we express the structure formula as:



From the chemical analysis given by Pask and Warner (1954) (see analysis A in Table 1 of their paper), the following formula is calculated:



The agreement is very good as regards the tetrahedral Al and the Mg, but less good as regards the  $\text{Fe}^{2+}$  and Al in octahedral coordination. Similar tests have been applied to three other chlorites namely:

Penninite, from Binnental (Switzerland), supplied by F. A. Bannister (British Museum), analyzed by R. F. Youell (University of Leeds).

Prochlorite, from Salcombe, Devon (England), supplied by C. E. Tilley (University of Cambridge), analyzed by H. C. G. Vincent. (*Geol. Mag.*, **75**, p. 503, 1938).

Daphnite, from Cornwall (England), supplied and analyzed by J. S. Webb.

The results of these four tests are indicated in Fig. 3, which shows on a triangular diagram the total octahedral compositions as calculated from the chemical analyses (black circles) and as derived from the x-ray data (shaded circles). The diagram is divided into areas according to

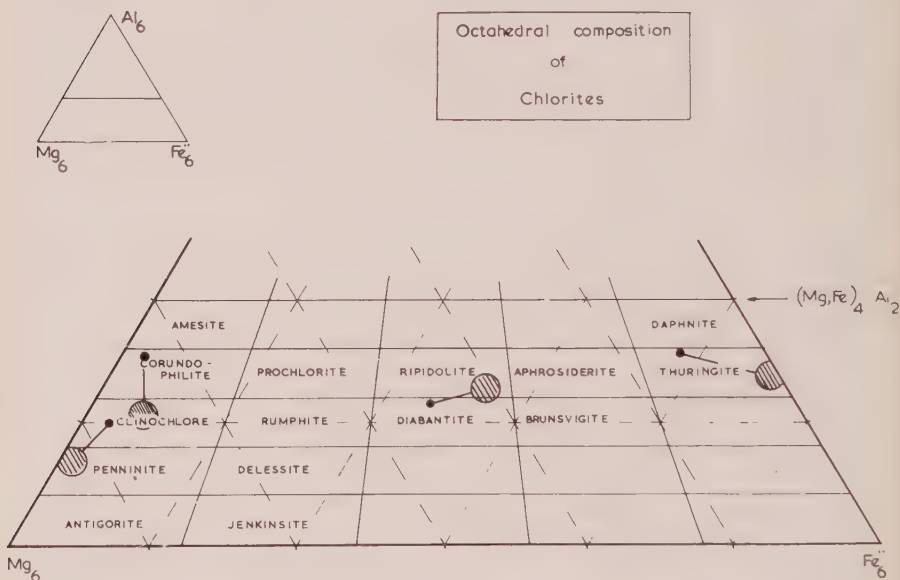


FIG. 3. A comparison of the octahedral compositions of four chlorites determined from chemical analyses (small black circles) and from x-ray data (large shaded circles). To avoid confusion, corresponding circles are joined.

Winchell's classification to show the general distribution of species names.

These results suggest that the identification of chlorite species from x-ray data can be carried out with a moderate degree of success.

#### ACKNOWLEDGMENTS

We are indebted to the Trustees of the British Museum, Dr. F. A. Bannister, Professor C. E. Tilley, Dr. J. A. Pask and Dr. J. S. Webb for gifts of specimens and to Mr. R. F. Youell for the chemical analysis of one specimen. One of us (F.H.G.) is indebted to the Department of Scientific and Industrial Research, London, for a maintenance grant, held during part of the period covered by this investigation, which was begun in the Physics Department of the University of Leeds, England, and continued at the Pennsylvania State University, U.S.A.



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## APPENDIX

*Conversion of reflected energy,  $E$ , to  $|F|^2$  for different experimental arrangements*

The increasing use by non-specialists of *x*-ray methods of identifying minerals, particularly by counter diffractometer methods, may justify the following notes. The basic formula (see, for example, James (1948), pp. 41-51) expresses the energy  $dE$  reflected by a small volume  $dv$  of non-absorbing crystal as it rotates within a beam of unpolarized *x*-rays of intensity  $I_0$  per unit cross-section as

$$dE = \text{Const. } I_0 |F|^2 \left( \frac{1 + \cos^2 2\theta}{\sin 2\theta} \right) dv \quad (1)$$

*Single crystals and oriented powder particles*

With micaceous minerals such as chlorites the particles may be well oriented with their basal planes parallel to a glass slide. Reflection from these planes will then be equivalent to reflection from an extended crystal face provided the orientation is sufficiently good.

*Case I. The incident x-ray beam falls wholly on an extended crystal face or an area of oriented powder particles*

Integration of equation (1), taking account of absorption in the specimen and assuming it to be sufficiently thick to transmit a negligible

portion of the radiation, leads to the expression

$$E = \text{Const.} \frac{I_0 S_0}{2\mu} |F|^2 \left( \frac{1 + \cos^2 2\theta}{\sin 2\theta} \right) \quad (2)$$

where  $\mu$  = linear absorption coefficient and  $S_0$  = cross-section of the incident beam where it falls on the specimen.

*Case II. The crystal or oriented powder specimen lies within the x-ray beam*

In mineral identification work a situation often arises where the individual crystal or oriented powder specimen lies wholly within the x-ray beam so that the fraction of the beam which can be reflected depends on the angular setting of the specimen within the beam. If the specimen area is  $A$ , it intercepts an area  $A \sin \theta$  of the beam and provided the beam is of uniform intensity the result is equivalent to equation (2) with  $A \sin \theta$  in place of  $S_0$ . The reflected energy is given by

$$E = \text{Const.} \frac{I_0 A}{2\mu} |F|^2 \sin \theta \left( \frac{1 + \cos^2 2\theta}{\sin 2\theta} \right) \quad (3)$$

It is assumed that the areal dimensions are large compared with the thickness so that "edge" effects are negligible.

*Cases I and II combined*

With counter diffractometers, it often happens that the amount of material available or that the size of the crystal flake is such that the incident radiation falls wholly on the specimen only when  $\theta$  is greater than a certain value. If  $W$  is the width of the specimen,  $\delta$  the divergence of the radiation, and  $R$  the spectrometer radius, then the limiting condition is  $R\delta = W \sin \theta$ . Equation (3) is appropriate when  $\sin \theta < R\delta/W$ , and equation (2) when  $\sin \theta > R\delta/W$ .

*Powdered specimens with random orientation*

For a small volume  $dv$  of randomly oriented powder particles having negligible absorption, the equation analogous to (1) is

$$dE = \text{Constant } p I_0 |F|^2 \cos \theta \left( \frac{1 + \cos^2 2\theta}{\sin 2\theta} \right) dv \quad (4)$$

where  $p$  is the multiplicity factor for the reflecting planes concerned, and  $dE$  is the energy per unit time in the whole diffracted halo.

*Case III. Counter diffractometer technique with a large area of randomly oriented powder particles*

By integration of equation (4) for a "thick" block of powder, assuming that the radiation falls wholly on the specimen and that the window



of the Geiger counter accepts a fixed length out of each diffracted halo, we obtain

$$E = \text{Const. } \phi I_0 S_0 |F|^2 \left( \frac{1 + \cos^2 2\theta}{\sin \theta \sin 2\theta} \right) \quad (5)$$

*Case IV. Counter spectrometer technique with small area of randomly oriented powder particles*

A specimen of area  $A$  intercepts an area  $A \sin \theta$  of the incident beam, so that  $A \sin \theta$  replaces  $S_0$  in equation (5) and we have

$$E = \text{Const. } I_0 A |F|^2 \left( \frac{1 + \cos^2 2\theta}{\sin 2\theta} \right) \quad (6)$$

*Powdered specimens with partial orientation*

A situation which is not amenable to precise calculation arises with imperfectly oriented powder specimens. Comparison of equations (2) and (5) relating to "large" specimens, with equations (3) and (6) relating to "small" specimens, shows that in passing from fully oriented reflecting planes to randomly oriented crystallites an additional factor of  $1/\sin \theta$  is introduced into the expressions for  $E$ . For a given set of reflecting planes  $\sin \theta$  is proportional to  $n$ , the order of the reflection in the Bragg equation. Thus for the two reflections (001) and (002), the ratio of their angular functions changes by a factor 2/1 depending on whether we assume a fully ordered or fully disordered arrangement of the reflecting particles. For higher orders, the ratio depends less critically on which formula is used. In the identification of chlorites, however, and in all similar problems, the lower orders are likely to be the most useful and therefore the question of orientation is an important one.

When dealing with flakey powders, such as commonly occur with micaceous minerals, it is recommended that identification procedures which involve intensity measurements should be carried out with (i) aggregate specimens oriented as well as possible by careful sedimentation, and (ii) powder specimens prepared with a minimum of orientation, e.g., by filling the specimen holder from the back rather than from the front surface.

*Other difficulties*

Under this heading brief reference may be made to other difficulties inherent in the precise interpretation of x-ray intensities. When single crystals and mineral flakes rather than fine powders are used, extinction effects may modify the reflected intensities, especially of strong, low-order reflections. When composite specimens containing several crystal-line components are used, questions of differential absorption arise par-

ticularly when the absorption coefficients of the components are very different. These difficulties are minimized by using fine powders. The formulae given for  $E$  refer to the total reflected energy whereas experimentally the peak intensity is often measured. The two are proportional for peaks of constant width. The present application is concerned mainly with low-order reflections of which the widths will be largely constant.

#### *Other techniques*

These notes have been written mainly in relation to Geiger counter diffractometer technique. When photographic technique is used, equation (3) is still valid for a small flake of crystal or a thin fragment of well-oriented powder rotated within an  $x$ -ray beam. For stationary powder blocks and powder rods, equations (5) and (6) are not valid and formulae (see James, 1948) must be used containing the appropriate absorption factors.

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*Manuscript received Jan. 21, 1955.*



# THE OCCURRENCE AND PROPERTIES OF METATYUYAMUNITE, $\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2 \cdot 3-5\text{H}_2\text{O}^*$

T. W. STERN, L. R. STIEFF, M. N. GIRHARD, AND ROBERT MEYROWITZ, *U. S. Geological Survey, Washington 25, D. C.*

## ABSTRACT

Metatyuyamunite  $[\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2 \cdot 3-5 \text{H}_2\text{O}]$ , a new mineral, is a dehydration product of tyuyamunite  $[\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2 \cdot 5-8.5 \text{H}_2\text{O}]$ . This lower hydrate of tyuyamunite has been collected from more than 35 localities on the Colorado Plateau and from several localities in Fall River County, S. Dak., and Campbell and Johnson counties, Wyo. The optical properties and specific gravity of metatyuyamunite and tyuyamunite are significantly different. Metatyuyamunite samples from two localities in Mesa County, Colo., have been chemically analyzed. The hydration of metatyuyamunite has been examined as a function of water vapor pressure. Plateaus on the dehydration curve are at values of 5 and 8.5 moles of water per mole of  $\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2$ . Metatyuyamunite was rehydrated to tyuyamunite by placing it in a moist atmosphere. The x-ray diffraction powder patterns of metatyuyamunite and tyuyamunite are distinctly different, indicating a change in phase. Base-exchange studies show that the metatyuyamunite structure may contain several per cent potassium. X-ray diffraction powder patterns of this base-exchanged product do not reveal the presence of carnotite  $[\text{K}_2(\text{UO}_2)_2(\text{VO}_4)_2 \cdot 1-3 \text{H}_2\text{O}]$ . Two samples of metatyuyamunite from Mesa County, Colo., are not in radium-uranium equilibrium, and their Ra/U ages are 25,000 and 150,000 years.

## INTRODUCTION

Variations in the water content of tyuyamunite have been studied by Hillebrand (1924, p. 202), Rohde (1925, p. 377), and Murata, Cisney, Stieff, and Zworykin (1951, p. 323). Merwin (1924, p. 210) found that the density of the mineral increases as it is dehydrated. An analogous variation in the water content of minerals of the torbernite and metatorbernite groups has been discussed in papers by Beintema (1938, p. 155), J. W. Frondel (1951, p. 249), C. Frondel (1951*a*, p. 678; 1951*b*, p. 681), Mrose (1950, p. 529), and Nuffield and Milne (1953, p. 476). The water content of sengierite (Vaes and Kerr, 1949, p. 109) should be studied when and if sufficient amounts of the mineral can be made available.

Both the torbernite group and synthetic  $\text{KUO}_2\text{VO}_4$  have a layer type structure (Beintema, 1938, p. 155; Sundberg and Sillen, 1950, p. 377). In  $\text{KUO}_2\text{VO}_4$ , sheets of  $(\text{UO}_2\text{VO}_4)^-$  are separated by  $\text{K}^+$  layers. Tyuyamunite  $\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2 \cdot 5-8.5\text{H}_2\text{O}$  has a layer structure with the metal-oxygen sheets bonded together by  $\text{Ca}^{++}$  ions. This layer structure is stable for values of  $n\text{H}_2\text{O}$  up to 8.5, and the water content ranges down to 3. The hydration-dehydration effect is reversible in the range  $3-8.5\text{H}_2\text{O}$ .

\* Publication authorized by the Director, U. S. Geological Survey.

Metatyuyamunite, a new mineral, contains 3.5 moles of water per mole of  $\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2$  and has distinct *x*-ray powder diffraction pattern and optical properties. Various hydrates within the range of stability have been found in nature, and unless specimens are collected in moisture-proof containers, they may dehydrate before they are examined in the laboratory. Crystal lattice studies of tyuyamunite, carnotite, and sengierite have been made by Donnay and Donnay (1954, p. 323).

#### OCCURRENCE OF METATYUYAMUNITE

The first specimen of metatyuyamunite examined in the present study was collected by L. R. Stieff in 1948, from an outcrop at the lower portal of the Jo Dandy mine, Montrose County, Colo. Purification of this sample for detailed mineralogic study was not possible because of the fine-grained nature of the metatyuyamunite and the presence of clay. Since 1948, metatyuyamunite has been collected from more than 35 localities on the Colorado Plateau and also from several localities in Fall River County, S. Dak., and in Campbell and Johnson counties, Wyo. The specimens from the Colorado Plateau occur in the Shinarump conglomerate (Triassic), Chinle formation (Triassic), Todilto limestone (Jurassic), and Salt Wash sandstone member of the Morrison formation (Jurassic). In most localities tyuyamunite and metatyuyamunite form pulverulent masses or are disseminated in sandstone or limestone. Tyuyamunite and metatyuyamunite are difficult to purify as they are commonly mixed with carnotite—the potassium uranyl vanadate—and other impurities.

Yellow crystals forming radial aggregates of metatyuyamunite from the Small Spot (pl. 1) and May Day mines, Calamity Mesa, Mesa County, Colo., have been studied in detail. The crystals, which are as long as 3 mm., filled fractures in the Salt Wash sandstone member of the Morrison formation (Jurassic). The metatyuyamunite formed on gypsum, which coated the fracture walls. In these samples the metatyuyamunite and tyuyamunite occurred in such a manner that by very careful work they could be hand-separated and purified.

#### PHYSICAL PROPERTIES

Metatyuyamunite is canary yellow to greenish yellow. It has perfect micaceous (001) cleavage. Cleavage on (010) and (100) is distinct. The crystals have an adamantine luster and massive material is distinctly waxy. Hardness is about 2 and the material is not brittle. The specific gravity of metatyuyamunite ranges from 3.8 to 3.9. On the hydration of metatyuyamunite to tyuyamunite the specific gravity drops to 3.6. Both tyuyamunite and metatyuyamunite fuse relatively easily, and this

property can be used to distinguish these minerals from carnotite when the samples contain only tyuyamunite or metatyuyamunite and carnotite. Both tyuyamunite and metatyuyamunite may turn yellow green on exposure to sunlight (Chirvinsky, 1924, p. 291).

#### OPTICAL PROPERTIES

Both tyuyamunite and metatyuyamunite are orthorhombic, biaxial negative, and occur as basal plates. These minerals are colorless to pale yellow in transmitted light and faintly pleochroic with X nearly colorless, Y very pale canary yellow, and Z pale canary yellow. X is



PLATE 1. Photomicrograph of bladed crystalline metatyuyamunite, Small Spot mine, Calamity Mesa, Mesa County, Colo.

perpendicular to the plate, and the plate is elongated in the Z direction (pl. 1). The optical orientation is the same as that given by Merwin (1924, p. 208) and Dolivo-Dobrovolskiy (1925, p. 367). The dispersion is  $r < v$  weak. The interference figure does not show strong enough dispersion to prove orthorhombic symmetry. The indices of refraction given in Table 1 were determined using arsenic tribromide, precipitated sulfur, and arsenic disulfide immersion liquids (Meyrowitz and Larsen, 1951, p. 746). The indices of refraction change significantly in air (Table 1) owing to the rapid hydration or dehydration of the material. Therefore, index of refraction determinations must be made immediately after making the slide.

Norman Herz, of the U. S. Geological Survey, determined the axial



TABLE 1. OPTICAL PROPERTIES OF TYUYAMUNITE AND METATYUYAMUNITE

Orientation		"Tyuyamunite"		Metatyuyamunite	
		(1)	(2)	(3)	(4)
<i>c</i>	$\alpha$	$1.72 \pm 0.01$	1.75 to 1.80 (calculated)	1.68 (calc.)	1.62 (calc.)
<i>a</i>	$\beta$	$1.868 \pm 0.005$	1.927 to 1.932	$1.835 \pm 0.002$	$1.842 \pm 0.002$
<i>b</i>	$\gamma$	$1.953 \pm 0.005$	1.965 to 1.968	$1.865 \pm 0.002$	$1.899 \pm 0.002$
2V		48°	45° to 51°	45°	48°

- (1) Sample from Henry Mountains, Utah (Ross, 1924).  $Y=b$  only optical orientation given.
- (2) Sample from Paradox Valley, Colo. (Merwin, 1924).
- (3) Sample from Small Spot mine, Mesa County, Colo. Measurements at conditions prevailing in Washington, D. C.
- (4) Sample from May Day mine, Mesa County, Colo. Measurements at conditions prevailing in Washington, D. C.

angle, 2V, with a universal stage.  $\alpha$  was calculated. The changes in the indices of refraction caused by the variation of water content are shown in Table 2. The indices of refraction for specimens from the Small Spot mine are given for material that was kept over water in a closed container for 16 days, with a water vapor pressure of about 16 mm. Hg (col. 1); for material that was stored for 16 days in a desiccator over concentrated  $H_2SO_4$ , which has a water vapor pressure of 0.6 mm. Hg (col. 2); and for material that was heated to 100° C. (col. 3). On dehydration the indices of refraction and 2V increase, but the optical orientation, dispersion, and pleochroism remain essentially the same.

TABLE 2. VARIATION IN OPTICAL PROPERTIES OF TYUYAMUNITE AND METATYUYAMUNITE FROM SMALL SPOT MINE CAUSED BY HYDRATION AND DEHYDRATION

Orientation		Hydrated sample (1)	Desiccated sample (2)	Sample heated to 100° C. (3)
<i>c</i>	$\alpha$	1.57 (calculated)	1.88 (calculated)	1.73 (calculated)
<i>a</i>	$\beta$	$1.805 \pm 0.002$	$1.959 \pm 0.002$	$1.960 \pm 0.002$
<i>b</i>	$\gamma$	$1.851 \pm 0.002$	$1.976 \pm 0.002$	$2.02 \pm 0.01$
2V		42°	47°	49°

#### CHEMICAL COMPOSITION

C. S. Annell of the Geological Survey made semiquantitative spectrographic analyses (Table 3) to guide the quantitative chemical analyses of the Small Spot and May Day materials (Table 4).

TABLE 3. SEMIQUANTITATIVE SPECTROGRAPHIC ANALYSES OF METATYUYAMUNITE IN PER CENT

Analyst: C. S. Annell, U. S. Geological Survey

Locality	More than 10	1-10	0.1-1.0	0.01-0.1
Small Spot mine	U	V Ca	Al	Si Co K Ba
May Day mine <sup>1</sup>	U	V Ca	Sr Al Ba	Si Fe

<sup>1</sup> This spectrogram was made in a region in which potassium could not be detected. A flame photometer analysis by C. A. Kinser, U. S. Geological Survey, indicated that potassium was present in the 1-10 per cent range.

TABLE 4. CHEMICAL ANALYSES OF TYUYAMUNITE AND METATYUYAMUNITE

	Tyuyamunite				Metatyuyamunite		Theoretical composition	
	(1)	(2)	(3)	(4)	(5)	(6)	Metatyuya- munite 3 H <sub>2</sub> O (7)	Tyuya- munite 8 H <sub>2</sub> O (8)
Na <sub>2</sub> O	0.02							
K <sub>2</sub> O	.18	0.5		2.89		3.10		
CaO	5.63	5.9	5.56	5.42	6.22	5.83	6.49	5.88
BaO	.54			.28		.30		
SrO				.08		.09		
MgO	.09							
CuO	.04							
UO <sub>3</sub>	55.91	57.7	60.00	57.68	66.80	62.12	66.21	59.95
V <sub>2</sub> O <sub>4</sub>			2.01	1.27	2.25	1.33		
V <sub>2</sub> O <sub>5</sub>	18.76	19.5	16.40	18.39	18.27	19.79	21.05	19.06
H <sub>2</sub> O	18.83	16.3	16.03	13.53	6.57	7.33	6.25	15.11
Rem.				0.10		.11		
Total	100.00	99.9	100.00	99.64	100.11	100.00	100.00	100.00
Sp. gr.			3.6		3.8	3.9		

(1) Calamity Creek, Mesa County, Colo. (Hillebrand, 1924, p. 213).

(2) Henry Mountains, Utah (Schaller, 1924).

(3) Small Spot mine, Mesa County, Colo. Recalculation of sample 5 based on change in water content following hydration to tyuyamunite. Analysts: A. M. Sherwood and R. G. Milkey.

(4) May Day mine, Mesa County, Colo. Analyst: A. M. Sherwood.

(5) Small Spot mine, Mesa County, Colo. Analyst: A. M. Sherwood.

(6) May Day mine, Mesa County, Colo. Recalculation of sample 4 based on change in water content following dehydration to metatyuyamunite. Analyst: A. M. Sherwood.

(7) Theoretical composition of metatyuyamunite with 3 moles of water  
[Ca(UO<sub>2</sub>)<sub>2</sub>(VO<sub>4</sub>)<sub>2</sub> · 3H<sub>2</sub>O].

(8) Theoretical composition of tyuyamunite with 8 moles of water  
[Ca(UO<sub>2</sub>)<sub>2</sub>(VO<sub>4</sub>)<sub>2</sub> · 8H<sub>2</sub>O].

Hydration of the Small Spot material increased its water content from 6.57 to 16.03 per cent and its x-ray diffraction powder pattern showed larger interplanar spacings. The 2.89 per cent potassium content in the May Day specimen does not seem to be due to admixed carnotite. This sample is discussed further in the section on base exchange.

#### WATER CONTENT

The number of moles of water as a function of the partial pressure of the water has been determined for the Small Spot material (Fig. 1).

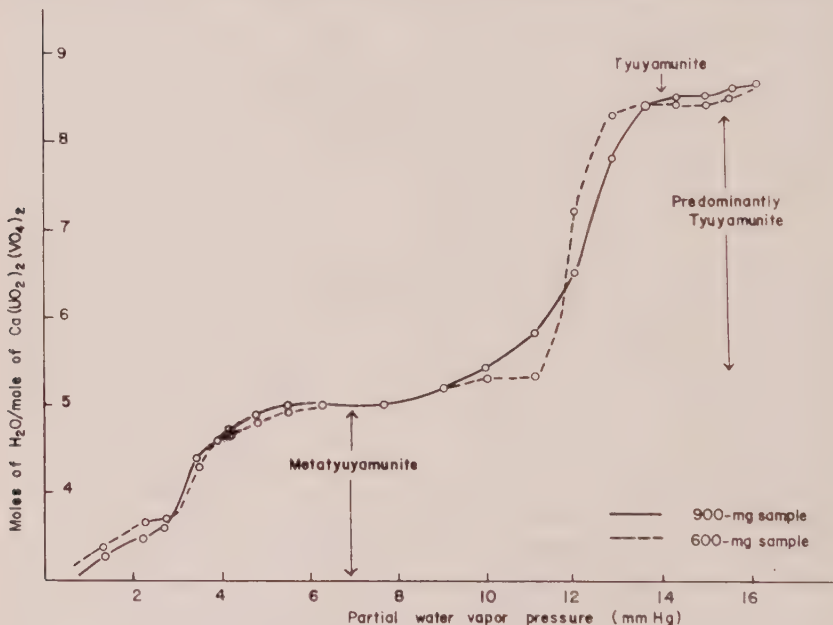


FIG. 1. Hydration of metatyuyamunite and tyuyamunite as a function of the partial pressure of water.

Two samples of the mineral weighing 600 and 900 mg. respectively were dehydrated in a desiccator containing concentrated sulfuric acid. The desiccator was kept in a constant temperature room (24° C.). All weighings were made in this room using a semimicrobalance. Equilibrium was assumed to have been reached when constant weight (within 0.2 to 0.3 mg.) was reached. The minimum interval between weighings was one day, and the sample was mixed thoroughly between weighings.

The partial pressure was varied by changing the concentration of aqueous sulfuric acid over which the sample was stored. The time required for equilibrium to be established at a new partial pressure varied



from 3 to 19 days. The partial pressure of the water vapor at each dilution was calculated from the approximate relative humidity of the sulfuric acid solutions using the table of Stevens (1916, p. 430), which shows the approximate relative humidity of dilute sulfuric acid solutions of various percentage compositions.

The water content of the dehydrated material was determined in the following manner. After the samples had come to equilibrium over concentrated sulfuric acid, a small portion of each sample was transferred to weighed porcelain microcrucibles. The crucible and contents were placed in the desiccator and weighed each day until the weight was constant. The weight of the crucible and contents was determined within 1 to 2 minutes after it was removed from the desiccator. The percentage of water was calculated from the loss on ignition. Several attempts were made to determine the water content by the Penfield method. These were not successful because the samples picked up water before the determination could be made.

#### X-RAY DIFFRACTION DATA

The plateaus in the dehydration curve at values of 5 and 8.5 moles of water per mole of  $\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2$  indicate that the structures of metatyuyamunite and tyuyamunite are complete. X-ray diffraction powder patterns for the two minerals are distinctly different (pl. 2 and Tables 5 and 6). Samples containing 3 to 5 moles of water per mole of  $\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2$  show slight differences among themselves in spacings due to changes in water content, but the spacings correspond essentially to those given for metatyuyamunite. Analogous differences exist in the tyuyamunite powder patterns in the range of 5 to 8.5 moles of water.

The differences in the x-ray diffraction powder patterns of tyuyamunite and metatyuyamunite are due to changes in the interlayer spacings between the  $(\text{UO}_2\text{VO}_4)^-$  sheets as the water content is changed either by hydration or by dehydration. Dehydrated specimens have smaller {001} interplanar spacings than do the more hydrated specimens. Murata, Cisney, Stieff, and Zworykin (1951) have already noted the correlation between water content and interlayer spacings.

George Ashby, of the Geological Survey, found that the space occupied by interlayer water in tyuyamunite is nearly the same as that occupied by interlayer water in montmorillonite. Tyuyamunite heated at 500° C. for 72 hours gives a single x-ray powder line in the low-angle region with an interplanar spacing about 5.8 Å. Assuming that 5.8 Å is the basal spacing of anhydrous tyuyamunite, the interlayer water space can be calculated as 2.7 Å for metatyuyamunite and 4.4 Å for tyuyamunite. The analogous water spaces for montmorillonite are 2.76 Å and 4.54 Å

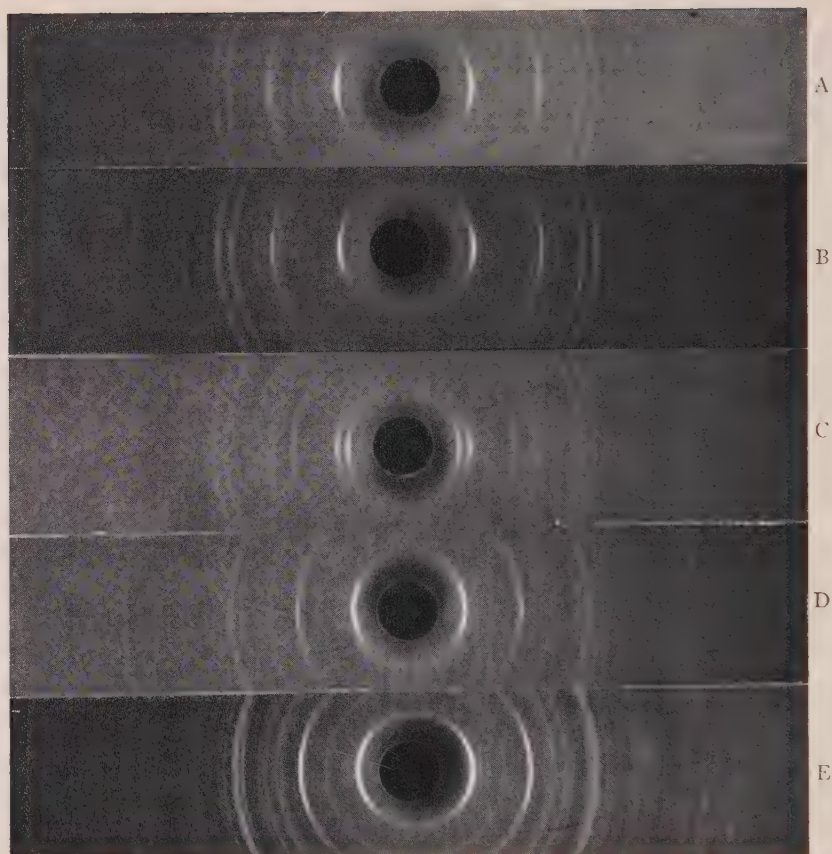


PLATE 2. X-ray diffraction powder patterns of metatyuyamunite and tyuyamunite.

- A. Metatyuyamunite from Small Spot mine, Mesa County, Colo. Sample in equilibrium with water vapor pressure of 0.6 mm. Hg. Film 5117.
- B. Metatyuyamunite from Small Spot mine, Mesa County, Colo. Sample in equilibrium with water vapor pressure of 4.8 mm. Hg. Film 5170.
- C. Metatyuyamunite and tyuyamunite from Small Spot mine, Mesa County, Colo. Note the two closely spaced lines at low angles of  $2\theta$ . Film 3342.
- D. Tyuyamunite from Small Spot mine, Mesa County, Colo. Sample in equilibrium with water vapor pressure of 15.1 mm. Hg. Film 3343.
- E. Tyuyamunite synthesized by K. J. Murata. Film 3336.

(Barshad, 1949, p. 675). These spacings suggest that metatyuyamunite has a single water layer and tyuyamunite has a double water layer.

In Table 5 the interplanar spacings are given for synthetic metatyuyamunite and metatyuyamunite from the Small Spot mine in equilibrium with a partial water vapor pressure of 0.6 mm. of Hg. Table 6 gives the

TABLE 5. X-RAY DIFFRACTION POWDER DATA FOR METATYUYAMUNITE  
(Cu/Ni RADIATION=1.5418 Å;  $d$  in Å)  
Data from D. D. Riska, U. S. Geological Survey

Synthetic metatyuyamunite (Film 109)		Metatyuyamunite $\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2 \cdot 3\text{H}_2\text{O}$ Small Spot mine, Mesa County, Colo. In equilibrium with water vapor pressure of 0.6 mm. Hg (Film 5117)	
$d_{\text{obs.}}$	$I^*$	$d_{\text{obs.}}$	$I^*$
9.94	vf		
8.51	vs	8.51	vs
6.51	w	6.66	f
5.19	w	5.22	w
4.48	f	4.48	f
4.22	s	4.22	s
3.77	w	3.77	w
3.55	f	3.55	f
3.24	mb	3.26	mb
3.04	mb	3.05	mb
2.85	vfb	2.81	f
2.72	f		
2.59	w	2.58	w
2.48	vfb		
2.19	fb		
2.13	wb	2.12	wb
2.05	f	2.05	f
2.01	f	1.99	f
1.96	f	1.95	f
		1.91	vfb
		1.88	vfb
		1.85	vfb
1.77	fb	1.77	fb
		1.73	vfb
1.69	vfb	1.69	vf
1.66	vfb	1.66	vf
		1.61	vfb
		1.57	vf
1.53	vf	1.53	vf
1.49	vf	1.50	vf

\* vs=very strong, s=strong, m=medium, w=weak, f=faint, vf=very faint, b=broad.

interplanar spacings of metatyuyamunite from the Small Spot mine in equilibrium with a partial water vapor pressure of 4.8 mm. of Hg and of tyuyamunite from the same mine with a partial water vapor pressure of 16.1 mm. of Hg. The patterns of metatyuyamunite ( $\sim 4.5 \text{ H}_2\text{O}$ ) and



TABLE 6. X-RAY DIFFRACTION POWER DATA FOR METATYUYAMUNITE AND  
TYUYAMUNITE (Cu/Ni RADIATION=1.5418;  $d$  IN Å)  
Data from D. D. Riska, U. S. Geological Survey

Metatyuyamunite $\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2 \cdot \sim 4.5 \text{ H}_2\text{O}$ . Small Spot mine, Mesa County, Colo. In Equilibrium with water vapor pressure of 4.8 mm. Hg. $a_0=10.63$ Å, $b_0=8.36$ Å, $c_0=16.96$ Å. (Film 5170)				Tyuyamunite $\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2 \cdot \sim 8.5 \text{ H}_2\text{O}$ . Small Spot mine, Mesa County, Colo. In equilibrium with water vapor pressure of 16.1 mm. Hg. $a_0=10.63$ Å, $b_0=8.36$ Å, $c_0=20.40$ Å. (Film 3343)			
$d_{\text{obs.}}$	$d_{\text{calc.}}$	$I^*$	$hkl$	$d_{\text{obs.}}$	$d_{\text{calc.}}$	$I^*$	$hkl$
8.55	8.48	vs	002	10.16	10.20	vs	002
6.55	6.57	f	110	6.51	6.57	f	110
5.25	$\begin{cases} 5.19 \\ 5.32 \end{cases}$	w	$\begin{cases} 112 \\ 200 \end{cases}$	5.28	5.31	vf	200
4.48	4.50	w	202	5.10	$\begin{cases} 5.10 \\ 5.14 \end{cases}$	m	$\begin{cases} 004 \\ 201 \end{cases}$
4.21	$\begin{cases} 4.18 \\ 4.24 \end{cases}$	s	$\begin{cases} 020 \\ 004 \end{cases}$	4.10	4.10	fb	212
3.77	$\begin{cases} 3.79 \\ 3.75 \end{cases}$	w	$\begin{cases} 121 \\ 022 \end{cases}$	3.40	3.40	mw	006
3.56	$\begin{cases} 3.56 \\ 3.54 \end{cases}$	f	$\begin{cases} 114 \\ 122 \end{cases}$	3.24	$\begin{cases} 3.24 \\ 3.24 \\ 3.23 \\ 3.26 \end{cases}$	mw	$\begin{cases} 221 \\ 205 \\ 024 \\ 310 \end{cases}$
3.25	$\begin{cases} 3.26 \\ 3.23 \end{cases}$	m	$\begin{cases} 310 \\ 221 \end{cases}$	3.03	$\begin{cases} 3.02 \\ 3.02 \end{cases}$	fb	$\begin{cases} 215 \\ 116 \end{cases}$
3.04	$\begin{cases} 3.04 \\ 3.06 \end{cases}$	m	$\begin{cases} 312 \\ 222 \end{cases}$	2.88	2.86	fb	206
2.82	$\begin{cases} 2.83 \\ 2.83 \\ 2.84 \\ 2.80 \end{cases}$	w	$\begin{cases} 006 \\ 313 \\ 223 \\ 024 \end{cases}$	2.65	$\begin{cases} 2.66 \\ 2.66 \\ 2.67 \\ 2.64 \\ 2.64 \end{cases}$	fb	$\begin{cases} 400 \\ 117 \\ 131 \\ 401 \\ 026 \end{cases}$
2.71	$\begin{cases} 2.70 \\ 2.71 \end{cases}$	f	$\begin{cases} 130 \\ 215 \end{cases}$	2.55	$\begin{cases} 2.55 \\ 2.55 \\ 2.55 \\ 2.56 \\ 2.56 \\ 2.57 \end{cases}$	fb	$\begin{cases} 008 \\ 315 \\ 207 \\ 225 \\ 126 \\ 402 \end{cases}$
2.58	$\begin{cases} 2.58 \\ 2.59 \\ 2.56 \\ 2.60 \\ 2.60 \\ 2.57 \end{cases}$	m	$\begin{cases} 322 \\ 314 \\ 125 \\ 224 \\ 116 \\ 132 \end{cases}$	2.17		fb	
				2.11		fb	

TABLE 6 (continued). X-RAY DIFFRACTION POWDER DATA FOR METATYUYAMUNITE AND TYUYAMUNITE (Cu/Ni RADIATION=1.5418;  $d$  IN Å)  
Data from D. D. Riska, U. S. Geological Survey

Metatyuyamunite $\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2 \cdot \sim 4.5 \text{ H}_2\text{O}$ . Small Spot Mine, Mesa County, Colo. In equilibrium with water vapor pressure of 4.8 mm. Hg. $a_0=10.63 \text{ Å}$ , $b_0=8.36 \text{ Å}$ , $c_0=16.96 \text{ Å}$ . (Film 5170)				Tyuyamunite $\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2 \cdot \sim 8.5 \text{ H}_2\text{O}$ . Small Spot mine, Mesa County, Colo. In equilibrium with water vapor pressure of 16.1 mm. Hg. $a_0=10.63 \text{ Å}$ , $b_0=8.36 \text{ Å}$ , $c_0=20.40 \text{ Å}$ . (Film 3343)			
$d_{\text{obs.}}$	$d_{\text{calc.}}$	$I^*$	$hkl$	$d_{\text{obs.}}$	$d_{\text{calc.}}$	$I^*$	$hkl$
2.49	$\begin{Bmatrix} 2.50 \\ 2.50 \end{Bmatrix}$	vf	$\begin{Bmatrix} 411 \\ 206 \end{Bmatrix}$	2.04		mw	
2.42		mb		1.98		vfb	
2.05		f		1.93		vfb	
2.00		w		1.86		vfb	
1.96		w		1.82		vfb	
1.88		f		1.76		vfb	
1.85		f		1.71		vfb	
1.77		f		1.69		vfb	
1.74		vf		1.66		vfb	
1.69		f		1.63		vfb	
1.66		f					
1.63		vf					
1.60		vf					
1.58		f					
1.53		f					
1.49		vfb					
1.48		f					

\* vs=very strong, s=strong, m=medium, mw=medium weak, w=weak, f=faint, vf=very faint, b=broad.

tyuyamunite ( $\sim 8.5 \text{ H}_2\text{O}$ ) have been indexed, and the powder patterns in Plate 2 show the change in the inner lines.

The single crystal data given by Donnay and Donnay (1954)—orthorhombic,  $a=10.63 \text{ \AA}$ ,  $b=8.36$ ,  $c=16.96$ ; pseudo *Pnan*—and referred to tyuyamunite are actually for the dehydrated form which all of us now call metatyuyamunite. We have used their data to index our

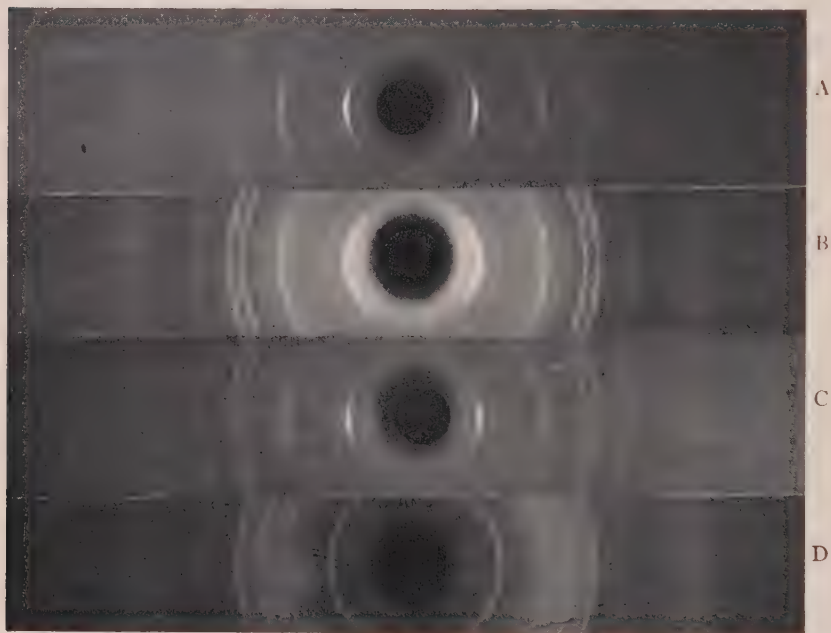


PLATE 3. X-ray diffraction powder patterns of metatyuyamunite, base-exchanged metatyuyamunite, and carnotite.

- A. Metatyuyamunite, Small Spot mine, Mesa County, Colo. In equilibrium with water vapor pressure of 0.6 mm. of Hg.; this material does not contain potassium.
- B. Metatyuyamunite, May Day mine, Mesa County, Colo. This material contains 2.89 per cent  $\text{K}_2\text{O}$ . Same hydration as A. Film 5219.
- C. Base-exchanged metatyuyamunite from Small Spot mine, Mesa County, Colo. Same hydration as A. Film 5220.
- D. Carnotite. Synthesized by K. J. Murata. Film 238.

pattern of metatyuyamunite. Our pattern of tyuyamunite has been indexed using the same  $a$  and  $b$  as above, but with  $c=20.40 \text{ \AA}$ , assuming an expansion in that direction.

#### BASE EXCHANGE

The metatyuyamunite from the May Day mine contains 2.89 per cent  $\text{K}_2\text{O}$ . These crystals are optically homogeneous, and no evidence of the



presence of carnotite could be found from *x*-ray examination of the material. To study the effect of the presence of  $K_2O$  in the material from the May Day mine, C. A. Kinser, of the Geological Survey, base-exchanged some of the  $CaO$  in the Small Spot sample with  $K_2O$ . A 100-mg. sample of metatyuyamunite was treated with 25 ml. of solution containing 3.0 mg. of  $K_2O$  as  $KCl$ . After base-exchanged reaction had taken place, the solution contained 0.80 mg.  $K_2O$  and 1.22 mg.  $CaO$ . The 2.20 mg. of  $K_2O$  that was removed from the solution by the reaction should liberate 1.30 mg. of  $CaO$ . The base-exchanged sample from the Small Spot mine, an untreated sample from the Small Spot mine, and the base-exchanged potassium-containing May Day mine material were stored in an atmosphere with a partial pressure of water of 1.2 mm. of Hg at  $24^\circ C$ . *X*-ray diffraction powder patterns were made of the three samples. For comparative purposes *x*-ray diffraction powder patterns are shown for metatyuyamunite from the Small Spot mine, metatyuyamunite from the May Day mine, base-exchanged metatyuyamunite, and carnotite in Plate 3. These experiments show that samples of metatyuyamunite can contain significant amounts of potassium. The transition point from orthorhombic symmetry of tyuyamunite to monoclinic symmetry of carnotite has not been determined. The base-exchange reaction easily converts tyuyamunite to carnotite, but the reaction from carnotite to tyuyamunite proceeds very slowly (Murata, Cisney, Stieff, and Zyorykin, 1951).

#### RADIUM-URANIUM AGE

Radium-uranium equilibrium is established when the rates of radium formation and decay are equal. Ninety-nine per cent of radium-uranium equilibrium will be attained in approximately 500,000 years (Knopf, 1931, p. 110). If minerals do not contain radium in proportion to the equilibrium amount of  $3.32 \times 10^{-7}$  g. Ra/g. U, this condition may be attributed to either one or both of the following causes:

1. The mineral may be less than 500,000 years old if the radiogenic lead content is very low.
2. The material may have been influenced by very recent selective leaching of radium or enrichment of uranium.

The metatyuyamunite samples from both the Small Spot mine and the May Day mine contain less than 0.001 g. Pb/g. sample. These well-crystallized specimens seem to be fresh and unaltered; therefore, the loss of large amounts of radium by selective leaching does not seem probable. For these reasons it is assumed that these samples can be dated approximately by the radium-uranium-equilibrium method.

Nuclear-track studies of both metatyuyamunite and carnotite showed

the absence of radiocolloids (local concentration of radium\* or other relatively short half-lived daughter products), which suggests that the minerals have not been recently leached of uranium with respect to radium. Furthermore, the number of alpha tracks associated with these minerals is small, relative to their uranium content, confirming a lack of radium-uranium equilibrium.

For the two metatyuyamunite specimens, Table 7 gives their uranium content, theoretical radium content assuming radioactive equilibrium, experimentally determined radium content, and age.

TABLE 7. RADIUM CONTENT, DEPARTURE FROM EQUILIBRIUM, AND RADIUM-URANIUM AGE OF METATYUYAMUNITE SPECIMENS FROM MESA COUNTY, COLO.

Location	Per cent U <sup>1</sup>	Theoretical g. Ra/g. sample <sup>2</sup>	Experimental g. Ra/g. sample <sup>3</sup>	Per cent equilibrium	Approximate age (years)
May Day mine, Mesa County, Colo.	52.00	$1.73 \times 10^{-7}$	$1.08 \times 10^{-7}$	62.4	150,000
Small Spot mine, Mesa County, Colo.	55.59	$1.84 \times 10^{-7}$	$0.333 \times 10^{-7}$	18.1	25,000

<sup>1</sup> Analyses by A. M. Sherwood and R. G. Milkey, U. S. Geological Survey.

<sup>2</sup> Computed from g. Ra/g. U in equilibrium =  $3.32 \times 10^{-7}$  g. Ra/g. U.

<sup>3</sup> Analyses by John Rosholt, U. S. Geological Survey.

Crystalline carnotite from the Bridger Jack Flat, Cane Springs Pass, Utah, is also of very recent geologic age. Using the data given by Knopf (1931, p. 351) and modern decay constants, the age of the carnotite from Bridger Jack Flat is approximately 170,000 years. Additional samples of crystalline carnotite from the Parco Mine, Yellow Cat group, Grand County, Utah, and from the Jo Dandy mine, Montrose County, Colo., have ages of 40,000 and 15,000 years, respectively, as determined by us in 1950. Thus, the age of some crystalline metatyuyamunite and carnotite specimens from the Colorado Plateau is Quaternary. These minerals have been formed from the vanadium-uranium deposits of the Plateau whose age has been determined as not older than Late Cretaceous or early Tertiary (Stieff and Stern, 1953).

#### ACKNOWLEDGMENTS

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# THE DISTRIBUTION AND IDENTIFICATION OF MIXED-LAYER CLAYS IN SEDIMENTARY ROCKS\*

CHARLES E. WEAVER

## ABSTRACT

Recent work has shown that randomly interstratified 2:1 clays are abundant in sedimentary rocks. X-ray diffraction patterns are presented which show a wide variety of randomly interstratified clays composed of illite-montmorillonite, chlorite-vermiculite, illite-chlorite-montmorillonite, and regularly interstratified chlorite-vermiculite. Methods of identifying and estimating the ratio of mixed layers are demonstrated.

## INTRODUCTION

Various authors have reported the occurrence of mixed-layer clays in relatively pure clay deposits and in soils. There is little, if any, literature concerning the abundance and varieties of mixed-layer clays occurring in shales, sandstones, and limestones. The author's investigation of the clay minerals in sedimentary rocks has shown that a large number of mixed-layer varieties are present, and that mixed-layer clays occur in abundance throughout the geologic section.

From an examination of over 6000 sedimentary rock samples from all over the United States, ranging in age from Cambrian to Recent, it is estimated that over 70 per cent of the samples contain some variety of mixed-layer clay.

Ability to recognize and identify mixed-layer clays will considerably increase the criteria used for the zonation and characterization of sedimentary rocks. It also appears likely that if minor environmental differences can be determined by clay mineral studies the mixed-layer clays hold the answer. It is more likely that mixed-layer illite-montmorillonite and chlorite-montmorillonite, rather than pure illite and chlorite clays, are formed by marine diagenesis.

The 2:1 clay minerals (illite, montmorillonite, chlorite, vermiculite) can be considered to be formed of similar flake-shaped layers. The type of cation which occurs between these layers determines the type of clay, i.e.  $K^+$  = illite,  $(Mg \cdot Al)(OH)_2^+$  = chlorite,  $Ca^{++}$ ,  $Na^+$ ,  $Mg^{++}$ , and  $H_2O$  = montmorillonite and vermiculite (vermiculite is considered to be a coarse-grained montmorillonite). A non-mixed-layer clay will contain the same type of cation between each layer; however, if 5 or 10 layers contain  $K^+$  in the interlayer position (illite) and another 10 or 15 contain  $Ca^{++}$  and  $H_2O$  (montmorillonite), etc., a mixed-layer clay is the result. Mixed-layer clays apparently can form in all possible combinations of

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different layers (illite-montmorillonite, illite-chlorite, chlorite-montmorillonite, illite-chlorite-montmorillonite).

Mixed-layer clays in most cases are derived from the degradation or aggradation of pre-existing clay minerals; i.e. during weathering,  $K^+$  can be removed from some biotite layers,  $Mg^{++}$  and  $H_2O$  deposited, and a mixed-layer biotite-vermiculite formed; or in sea water the  $Ca^{++}$  and  $H_2O$  between some montmorillonite layers can be replaced by  $K^+$ , and a mixed-layer illite-montmorillonite formed.  $Mg(OH)_2$  also readily replaces  $Ca^{++}$  and  $H_2O$  and forms mixed-layer chlorite-montmorillonite.

It is difficult, at the present time, to determine if a mixed-layer clay in a sediment was formed by diagenesis in the basin of deposition or by weathering in the source area. However, it is believed that originally a large part of the mixed-layer illite-montmorillonite and chlorite-montmorillonite clays were formed by marine diagenesis, and mixed-layer chlorite-vermiculite clays, primarily because of their relatively large grain size, are usually considered to have been formed by weathering.

#### IDENTIFICATION

Monomineralic clays produce an  $(00l)$  series of  $x$ -ray reflections which are integral sequences of the  $(001)$  reflection. The  $(001)$  reflection is a measure of the clay thickness, e.g.  $10 \text{ \AA}$  for illite. Thus, the  $(00l)$  series for illite would be:  $(001) = 10 \text{ \AA}$ ,  $(002) = 5 \text{ \AA}$ ,  $(003) = 3.3 \text{ \AA}$ ,  $(004) = 2.5 \text{ \AA}$ ,  $(005) = 2 \text{ \AA}$ , etc.

When the two different types of layers which form a mixed-layer clay occur in random intergrowth the clay is referred to as a random mixed-layer clay. In this case the  $(00l)$  series is not an integral sequence.

The  $(00l)$  values are average values resulting from the simultaneous scattering by both types of layers. This can be seen more clearly in Fig. 1.

The vertical lines in the diagram below each  $x$ -ray diffraction pattern show the position of the  $(00l)$  reflections for each type of layer. The longer lines show the position of the  $10\text{-\AA}$   $(00l)$  series (10, 5, 3.3, 2) and the shorter lines the position of the  $(00l)$  reflections of the expandable layers (12.4, 6.2, 4.13, 3.10, 2.07). The dark horizontal line indicates which adjacent reflections combine to produce the observed average reflection. These average reflections are referred to as  $(001)/(001)$ ,  $(002)/(003)$ , etc. indicating which two values combine to form the average value. The precise position will depend upon the relative amounts of the two layers, the form factor, and particularly in the case of the first maximum, the polarization factor. Actually, averages are effective between every pair of adjacent specific positions, but the intensity and sharpness of maxima are directly proportional to proximity. In those instances in which a node for one dominant species is flanked on both sides by equally

removed nodes for the minor component, the observed maximum merely exhibits minor broadening toward both sides. (See Fig. 1: 1 at 5.02 Å, or 1, 3 at 2.0 Å).

The theoretical basis for the identification of mixed-layer clay minerals

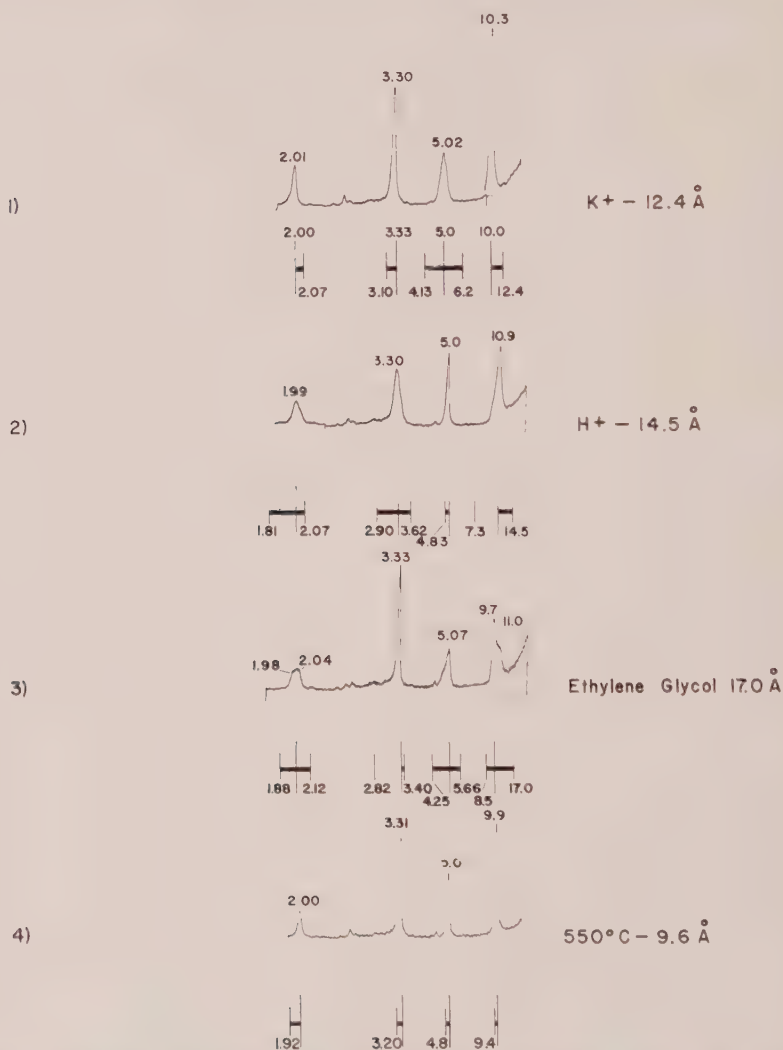


FIG. 1. Randomly interstratified clay (20% expanded layers/80% nonexpanded layers).

has been developed in papers by Hendricks and Teller (1942), Bradley (1945, 1950, 1953), MacEwan (1949), and Brown and MacEwan (1950).

Figure 1 contains x-ray diffraction curves of a clay composed of randomly interstratified nonexpanded 10-Å layers (illite) and 12.4-Å and

14.5-Å expanded layers (montmorillonite). When the exchangeable cations on the expanded layers are Na or K, the expanded layers contain only one layer of water and are approximately 12.4 Å thick. The (001)/(001) spacing occurs at 10.3 Å, the (002)/(002) at 5.02, and the (003)/(004) at 3.30. The 10.3-Å (001)/(001) spacing is an average obtained from the 10-Å and 12.4-Å reflection. The (002)/(002) value in curve 1, Fig. 1, lies between the (002) 6.2-Å reflection and the (003) 4.13-Å reflection and has a maximum near the (002) 5-Å maximum.

In the H<sup>+</sup> treated sample the expandable layers contain two layers of water and are approximately 14.5 Å thick. The first-, third-, and fifth-order peaks are broader and decreased in intensity as compared to the Na<sup>+</sup> sample, whereas the second-order peak is sharpened and increased in intensity. The broadening is caused by (00*l*) values which form the first-, third-, and fifth-order values moving farther apart; the sharpening by the (00*l*) values which form the second-order peak coming closer together.

In glycolated specimens, both the (001)/(001) and the (001)/(002) proximities are sufficient that they furnish observable maxima. The first is diffused at about 11 Å and the second much sharper at about 9.7. The (003) and (005) reflections nearly coincide, and the increased intensity of the 3.3-Å reflection is quite marked.

When the mixed-layer clay is heated to 550° C. the expanded layers collapse to a thickness of from 9.4 Å to 9.8 Å, depending upon the type of exchange cation. The resulting reflections are average values formed by adjacent (00*l*) values of the 10-Å layer and 9.4-Å to 9.8-Å layers.

In an untreated mixed-layer illite-montmorillonite clay it is often possible to tell whether the expanded layers have one or two layers of water by observing the relative sharpness and intensities of the peaks and by observing on which side of the 5-Å and 3.3-Å position the peaks lie. The reason for this can be seen in the diagrams in Fig. 1 which show the relative positions of the (00*l*) reflections.

Brown and MacEwan (1950) have computed theoretical curves for random mixtures of 10/12.4-Å, 10/14-Å, 10/15.4-Å, 10/17.7-Å, and 7.14/10-Å layers. With their formula, theoretical scattering curves were computed for the (001)/(001) reflection of randomly interstratified 10-Å and 17-Å layers at intervals of 10 per cent.

$$I = F_e^2 \Xi \frac{2f(1-f) \sin^2 \pi \frac{d_2 - d_1}{d'}}{1 - 2f(1-f) \sin^2 \pi \frac{d_2 - d_1}{d'} - f \cos 2\pi \frac{d_1}{d'} - (1-f) \cos 2\pi \frac{d_2}{d'}}$$

$f$  = proportion (expressed as a decimal fraction) of layers with higher spacing.

$d'$  = "apparent spacing" as measured on the film.

$d_1$  = higher interlayer spacing.

$d_2$  = lower interlayer spacing.



The form factor,  $F$ , and the polarization factor,  $\Xi$ , were not computed but were read from Bradley's (1945) experimentally estimated form factor curve which was compiled from various montmorillonite complexes.

Figure 2 is a curve showing the migration of this (001)/(001) peak as the percentage of expanded 17-Å layers is varied. The (001)/(001) was not calculated below about 30 per cent expanded layers. At 30 per

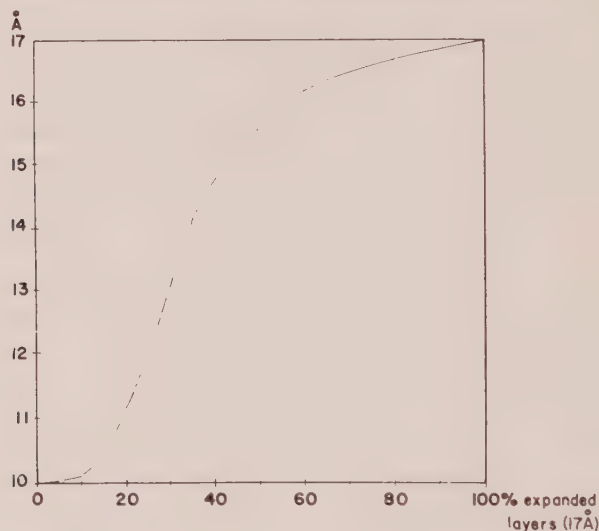


FIG. 2—Curve showing migration of (001)/(001) peak of randomly interstratified 10Å and 17Å layers.

cent the peak is very weak but increases as the percentage of expanded layers increases.

#### EXAMPLES OF RANDOMLY INTERSTRATIFIED ILLITE-MONTMORILLONITE CLAYS OCCURRING IN SHALES

Examination of a large number of shale samples indicates that the randomly interstratified expanded and nonexpanded layers occur in all possible ratios. Figures 3 and 4 contain a sequence of x-ray diffraction curves of mixed-layer clays in which the expanded layers range, at intervals of approximately 10 per cent, from less than 10 per cent to 80 per cent. Curves are given for mixtures of nonexpanded 10-Å layers and expanded 12.4-Å layers, and for 10-Å layers and 17-Å glycolated layers. In addition to a mixed-layer clay, kaolinite, chlorite, illite, and quartz are present in many of these samples.

In some cases the ratio of layers determined from the 10/12.4-Å curve

and the 10/17-Å do not show exact agreement. In most cases this discrepancy may be caused by the presence of a few intergrown chlorite layers, as the measured 10/12.4-Å value is usually too high and the 10/17-Å value too low. Heat treatment indicates that the chlorite layers are relatively scarce. Table 1 lists the (001)/(001) values obtained from

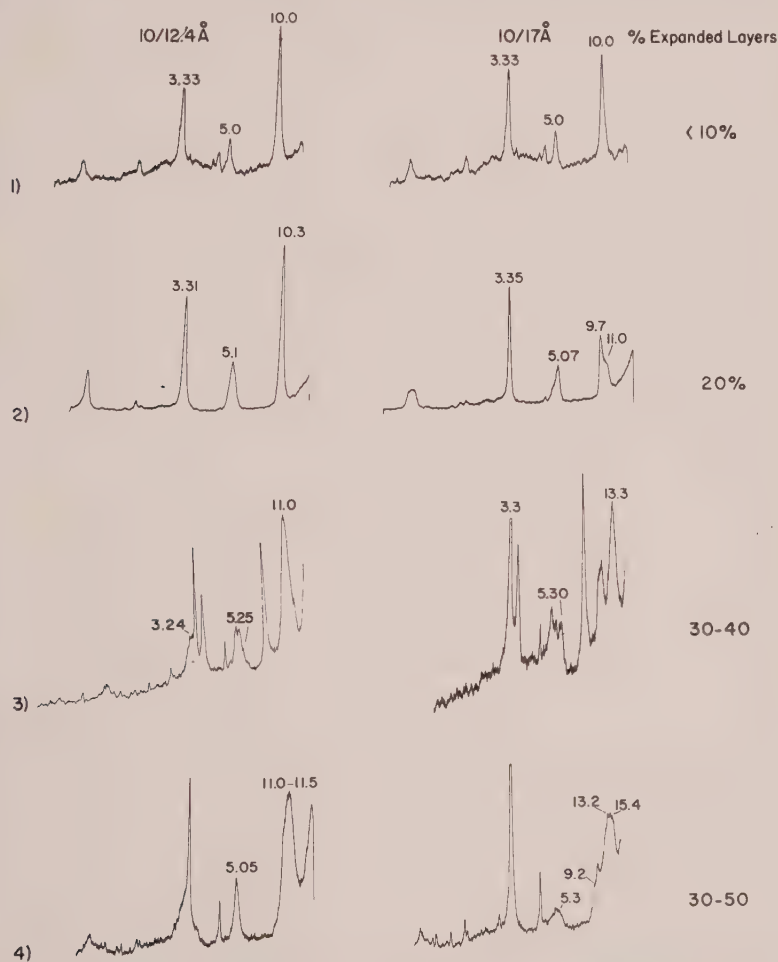


FIG. 3. Sequence of randomly interstratified clays (expanded and nonexpanded layers).

the curves in Figs. 3 and 4 and the calculated values taken from the curve, in Fig. 2 (10-Å/17-Å) and from Brown and MacEwan (10-Å/12.4-Å).

Curve 1 on Fig. 3 is an example of a mixed-layer clay which contains less than 10 per cent of expanded layers. When the clay is glycolated

there is no shifting of the peaks but there is a relative increase in the intensity of the (003)/(005), 3.33-Å peak as the (005) reflection of the expanded layers is moved closer to the (003) reflection of the 10-Å layers.

When the expanded layers are increased to 20 per cent the reflection resulting from the mixture of 10-Å and 12.4-Å layers has moved to 10.3 Å. When this material is glycolated, two high-angle peaks are formed, 11.0 Å and 9.7 Å, one resulting from a 10-Å/17-Å combination and the other from a 10-Å/8.5-Å combination. The combination 10-Å/17-Å peak gradually approaches a 17-Å value and the 10-Å/8.5-Å peak approaches an 8.5-Å value as the percentage of expanded layers increases, though there is not a straight-line relation as can be seen from the meas-

TABLE 1

Per cent Expanded Layers	10/12.4 Å		10/17 Å	
	Calculated (Å)	Measured (Å)	Calculated (Å)	Measured (Å)
<10	10	10	—	10
20	10.3	10.3	—	11.0
30	10.7	11.0	12.9	13.3
40	11.0	11.2	14.7	14.3
50	11.3	11.4	15.4	15.4
50	11.7	11.6	16.2	16.0
70	12.0	12.0	16.5	16.4
80	12.1	12.1	16.7	16.7

ured values and from the calculated curve. Brown and MacEwan (1950) have shown that when the two interfering reflections are relatively close, 10 Å and 12.4 Å, the calculated curve is nearly a straight line, and as the reflections move farther apart, the curve becomes more S shaped.

At a concentration of 30 to 40 per cent expanded layers the combination 5.0-Å/6.2-Å peak of the glycolated material increases suddenly from around 5.0 Å to 5.30 Å and continues increasing to a value of 5.6 Å, when 80 to 90 per cent of the layers are expanded. The 3.33-Å/3.40-Å peak approaches a 3.40-Å position as the per cents of expanded layers increase. This peak is obscured by the 3.35-Å quartz peak, but the asymmetrical shape of the 3.35-Å peak indicates that the mixed-layer clay has a peak near to 3.40 Å.

The composite 10-Å/12.4-Å peak approaches a 12.4-Å value as the proportion of expanded layers increases. The 5.0-Å/6.2-Å peak remains

near 5.0 Å throughout this range of values. This peak is quite weak and, if present, is probably obscured by the 5.0-Å peak of the illite, which is present in most of the samples. The 3.33-Å/3.10-Å peak decreases from a

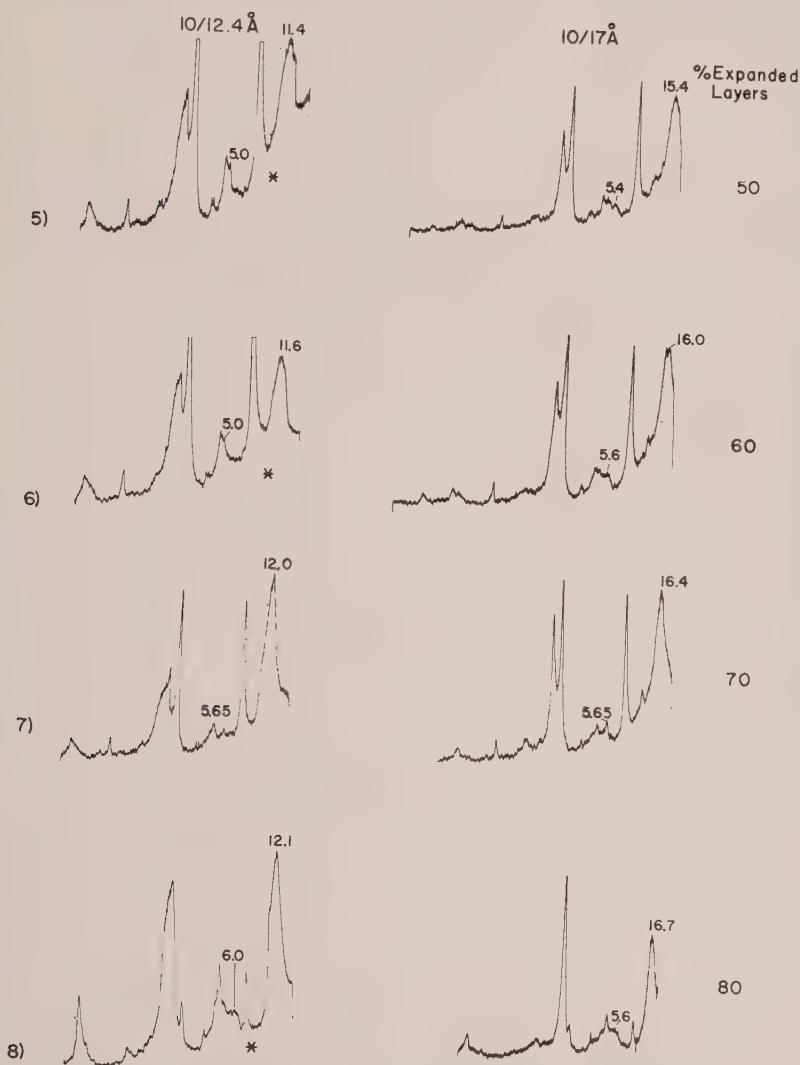


FIG. 4. Sequence of randomly interstratified clays (expanded and nonexpanded layers).

value of 3.31 Å to 3.24 Å as the percentage of expanded layers increases from 20 per cent to 30 per cent. Thereafter the decrease is less rapid.

Curve 4, Fig. 3, has a broad (001)/(001) peak for both the 10/12.4-Å



curve and the 10/17-Å curve. This sample appears to contain mixed-layer crystallites in which the percentage of expanded layers ranges from 30 to 50. Méring (1949) has shown that the smaller the number of unit layers in each crystallite, the broader the diffraction maxima. However, in this case it appears that the broad maximum is due to the presence of

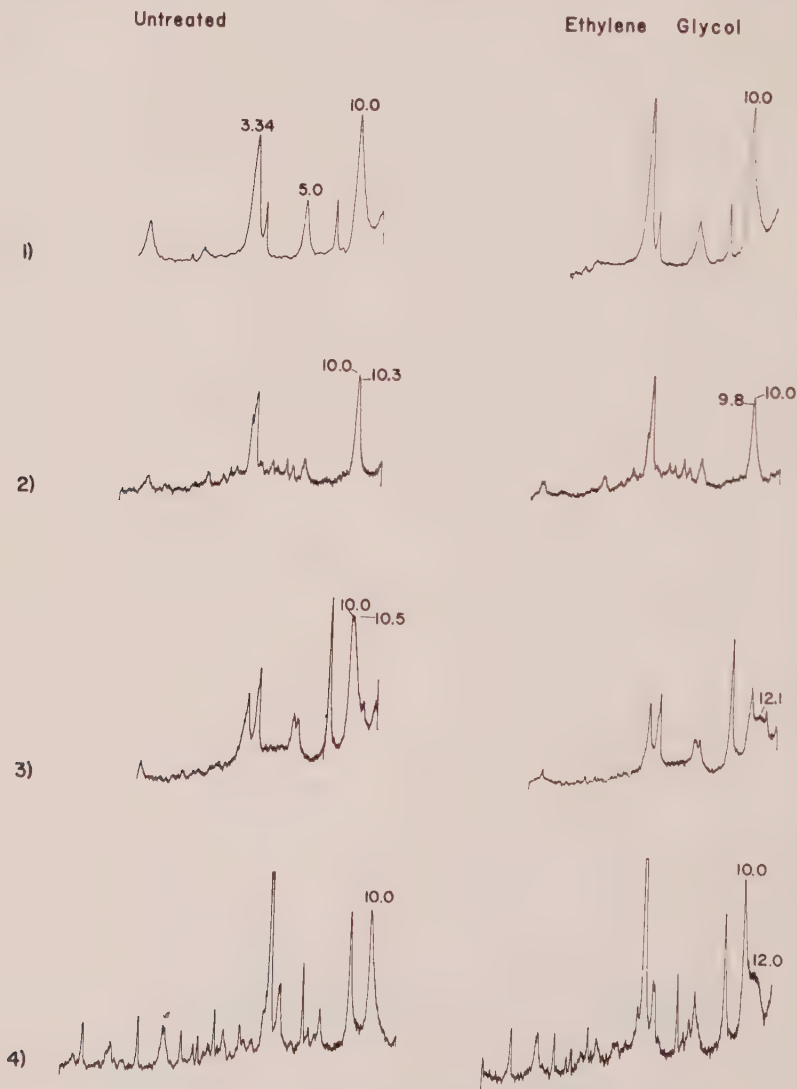


FIG. 5. Occurrence of mixed-layer clays with other clays (mixed layers—10–20% expanded/80–90% nonexpanded).

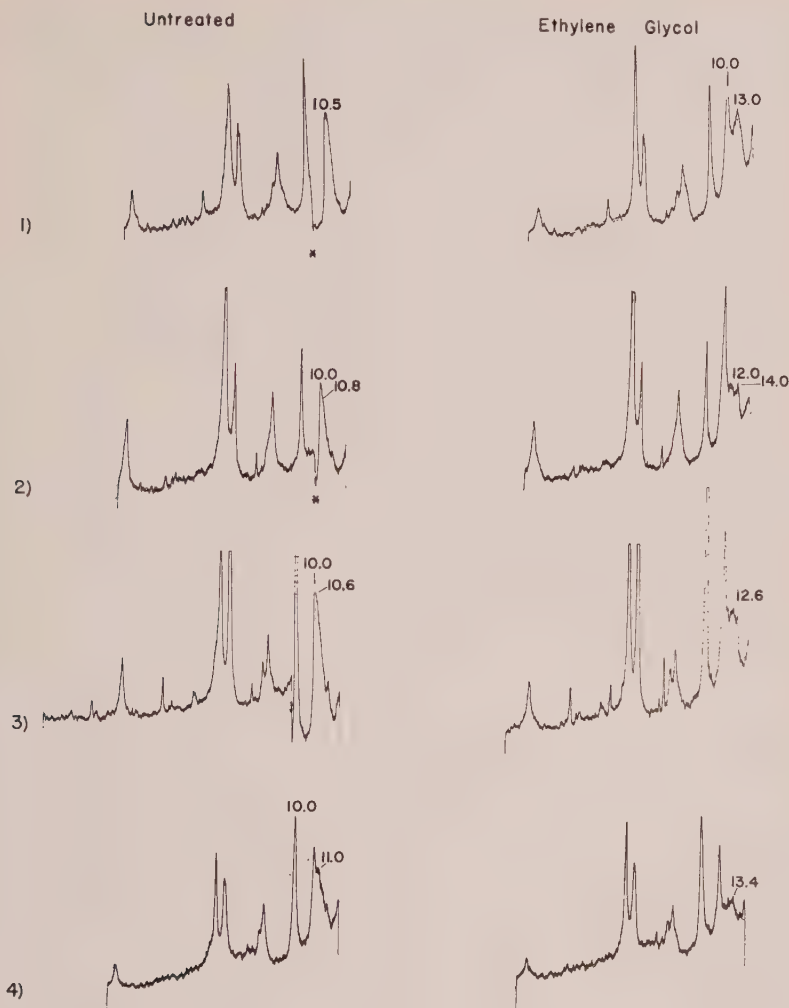


FIG. 6. Occurrence of mixed layer clays with other clays (mixed layers—20–30% expanded/70–80% nonexpanded).

crystallites with varying percentages of expanded layers rather than to the smallness of the crystallites.

Figure 4\* contains curves for 50, 60, 70, and 80 per cent expanded layers. It should be noted that for clays containing 60 to 100 per cent ex-

\* In the following figures an asterisk (\*) at the bottom of an x-ray diffraction curve indicates that the peak to the right of the asterisk is run at a setting having only one-half the sensitivity as the setting used in recording the curve to the left.

panded layers, there is only a 1-Å shift of the (001)/(001), 10/17-Å maxima, whereas there is a 6-Å shift in the interval from 10 to 60 per cent expanded layers.

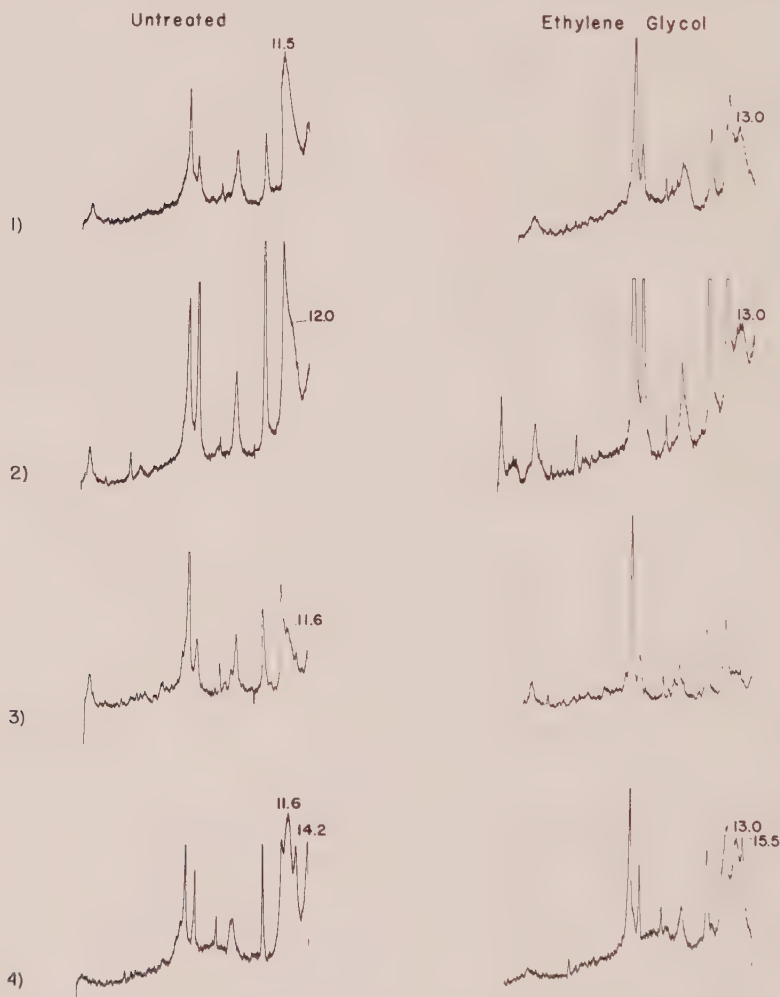


FIG. 7. Occurrence of mixed-layer clays with other clays (mixed layers—30% expanded/70% nonexpanded).

Figures 5, 6, 7, and 8 contain x-ray diffraction curves of the less-than-two-micron fraction of a variety of shales and sandstones. These curves illustrate a variety of ways in which the mixed-layer clays just discussed can combine with other clays to complicate the interpretation of the x-ray curves.

Figure 5 illustrates the case where a 10-Å illite and a 10.1-Å to 10.5-Å mixed-layer clay (10 per cent to 20 per cent expanded layers) occur together. In the first glycolated curve the presence of the mixed-layer clay

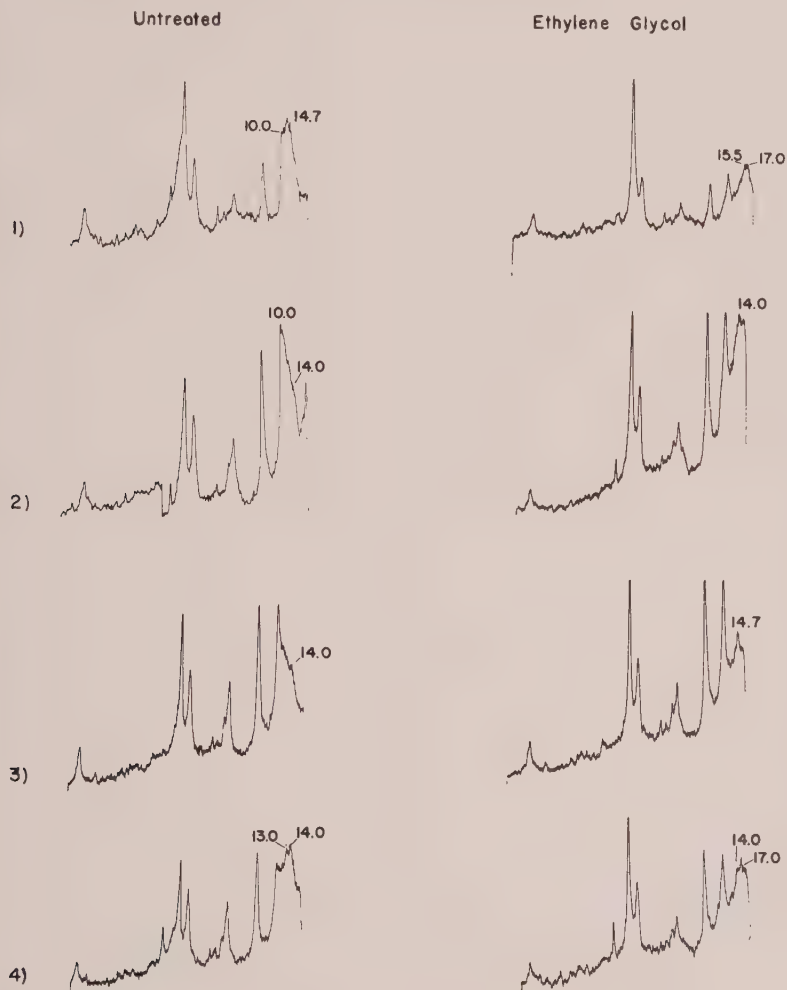


FIG. 8. Occurrence of mixed-layer clays with other clays (mixed layers—>40% expanded/<60% nonexpanded).

is shown only by a slight sharpening of the first- and third-order peaks and an increase in intensity of the third order. In the second curve glycolation has caused the development of a more symmetrical first-order peak and formed a minor peak at 9.8 Å. In the third curve in which the first peak of the untreated sample has a broad 10.0-Å to 10.5 Å top, the



mixed-layer phase is quite obvious in the glycolated sample. The 10-Å peak of the fourth curve suggests the presence of normal illite clay; however, the glycolated sample shows the presence of considerable mixed-layer material.

The  $x$ -ray patterns in Fig. 6 illustrate a variety of ways in which 10-Å illite and 10.5-Å to 11.0-Å mixed-layer clays (20 to 30 per cent expanded layers) can occur. In all cases the presence of the mixed-layer clays becomes evident when a glycolated sample is  $x$ -rayed.

Figure 7 shows typical combinations of 10-Å illite and 11.5-Å to 12.0-Å mixed-layer material (30 per cent expanded layers). The mixed-layer clays in the unglycolated patterns are largely in the 10/14-Å form rather than the 10/12-Å. Curve 4 shows, in addition to a mixed-layer illite-montmorillonite (13 Å), a mixed-layer chlorite-vermiculite reflection (15.5 Å).

Figure 8 shows patterns resulting from mixtures of 10-Å illite peaks and broad 10-Å to 14-Å mixed-layer peaks (40 to 100 per cent expanded layers). In the glycolated patterns the high-angle mixed-layer peak is frequently obscured by the 14-Å chlorite peak.

$X$ -ray patterns of over 6000 sediments have revealed that approximately 50 per cent of them have mixed-layer clays of the types shown in the previous figures.

#### EXAMPLES OF RANDOMLY INTERSTRATIFIED CHLORITE-VERMICULITE

Figure 9 contains  $x$ -ray patterns of randomly interstratified chlorite and vermiculite layers. Chlorite has strong second-order (7.1-Å) and fourth-order (3.55-Å) reflections and weak first-order (14.2-Å) and third-order (4.73-Å) reflections. In vermiculite the interlayer matter ( $Mg^{++}$  and  $H_2O$ ) is less dense than in chlorite [ $Mg(OH)_2$ ] and as a result the first-order reflection (14.2 Å) is strong and the second and third orders are weak. Thus, in a mixed-layer chlorite-vermiculite clay the relative intensities of the 14.2-Å and 7.1-Å peaks can be used to estimate qualitatively the relative percentage of chlorite and vermiculite. However, as the relative height of the 7.1-Å and 14.2-Å chlorite peaks vary with chemical composition, quantitative evaluation would be complex.

The first three curves in Fig. 9 are  $x$ -ray patterns of treated bulk samples of randomly interstratified vermiculite-chlorite layers. The presence of such a mixture is suggested in the first curve by the relative intensities of the 14.0-Å and 7.2-Å peaks. When the sample is heated to 400° C., approximately 50 per cent of the layers collapse to 10 Å and the resulting  $x$ -ray curve has peaks at 12.6 Å (10-Å/14.2-Å), 8.0 Å (7.1-Å/10-Å), 4.90 Å (4.73-Å/5.0-Å), and 3.49 Å (3.33-Å/3.55-Å) which are average values obtained by interaction of the 14.2-Å (00 $l$ ) series and the

10-Å (00 $l$ ) series. When the sample is heated to 550° C. and the OH<sup>-</sup> is removed from the chlorite interlayer positions the second-, third-, and fourth-order chlorite reflections are destroyed and the curve contains only an 11.6-Å reflection formed by the 10-Å and 13.8-Å (with the loss

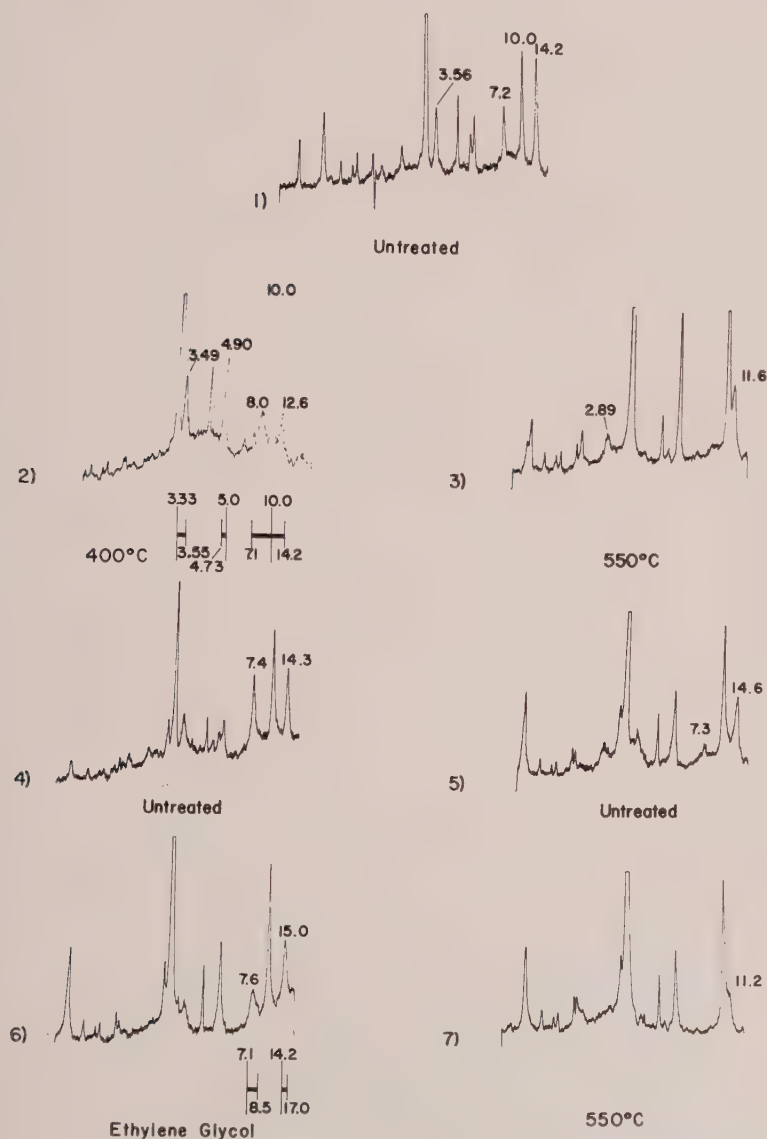


FIG. 9. Randomly interstratified chlorite/vermiculite layers.

of  $\text{OH}^-$  the chlorite shrinks from 14.2 Å to approximately 13.8 Å), (001) values and a 2.89-Å reflection (003)/(005).

Curve 4 shows a mixed-layer chlorite-vermiculite in which the chlorite is more abundant than in curve 1. The mixed-layer clay illustrated by curve 5 consists almost entirely of vermiculite layers. In the pattern of the glycolated sample it can be seen that the (001)/(001) peak has shifted to 15 Å and the (002)/(002) peak to 7.6 Å.

When the sample is heated to 550° C. and the expanded layer collapses to 10 Å the (001)/(001) peaks shift to a lower spacing of 11.2 Å.

### THREE-COMPONENT MIXED-LAYER CLAY MINERALS

Mixed-layer clays containing three components—illite, chlorite, and montmorillonite—are fairly common in shales. Figure 10 contains x-ray

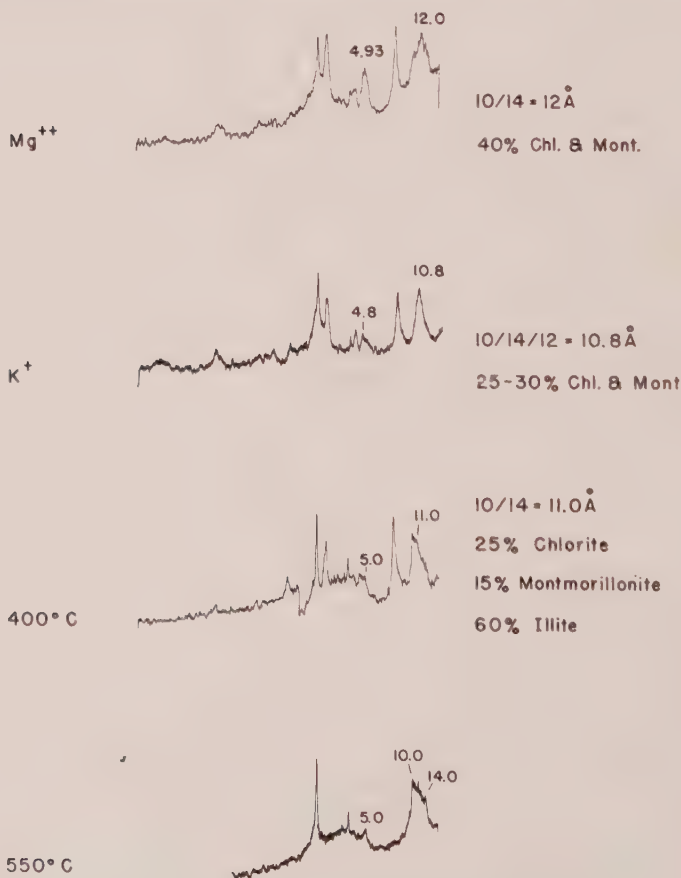


FIG. 10. Mixed-layer clay (illite/chlorite/montmorillonite).

diffraction curves of a typical clay of this type. Although interpretation of the data is rendered difficult by the presence of some discrete chlorite and illite, the following observations appear justified: The presence of chlorite layers is shown by the curves of the heated samples. The curve of the 400° C. treated clay has only collapsed to 11 Å and the 550° C. curve actually shows an increase in intensity as OH<sup>-</sup> is removed from the brucite layer of the chlorite.

When the mixed-layer clay is leached with MgCl<sub>2</sub>, the expanded layers absorb two layers of water and are approximately 14 Å thick. The form factor for chlorite is sufficiently similar to that for montmorillonite (Bradley, 1953), that the illite (10 Å)-chlorite (14 Å)-montmorillonite (14 Å) mixed-layer clay can be treated as a two-component, 10-Å and 14-Å system. The curve of the Mg<sup>++</sup> treated material indicates a mixture of approximately 40 per cent 14-Å chlorite-montmorillonite layers and 60 per cent 10-Å illite layers. Leaching the material with KOH collapses the expanded layers to 12 Å and lowers the (001)/(001) peak to 10.8 Å. The curve of the K<sup>+</sup> treated clay indicates that the mixed-layer clay has between 25 per cent and 30 per cent of layers greater than 10 Å. This lower value may be caused by the K<sup>+</sup> collapsing some of the expanded layers to 10 Å.

When the sample is heated to 400° C. the expanded montmorillonite layers collapse to 10 Å [illite(10 Å)-chlorite(14 Å)-montmorillonite (10 Å)] and a different two-component system, 10-Å and 14-Å layers, is formed. The 10-Å/14-Å peak at approximately 11 Å indicates a mixture of 75 per cent 10-Å illite-montmorillonite and 25 per cent 14-Å chlorite material. The mixed-layer clay is therefore composed of approximately 60 per cent illite layers, 25 per cent chlorite layers, and 15 per cent montmorillonite layers.

#### REGULAR MIXED-LAYER CLAYS

Regular mixed-layer clays are those in which two or more different types of layers occur in a regular sequence, i.e., AB, AB, AB, or AAB, AAB, etc. (Bradley, 1950 and 1953).

The (001) value of a regular mixed-layer clay is equal to the total thickness of the two or more types of layers which are present, i.e., the (001) value of a regular mixed-layer illite (10 Å) and montmorillonite (14 Å) would be 24 Å. Regular mixed-layer clays are differentiated from random mixed-layer clays in that the (001) series occurs in an integral sequence. It has been pointed out by Bradley (personal communication) that large (001) values (24 to 30 Å) are, in themselves, not proof of regularity. In random mixed-layer clays where the form factors are sufficiently different, in addition to an (001)/(001) average reflection, a low-angle



reflection will occur in the 24-Å to 30-Å region because the contributions from the layers with the higher scattering power are not completely canceled by those of the lower power. For example, in a random mixed-

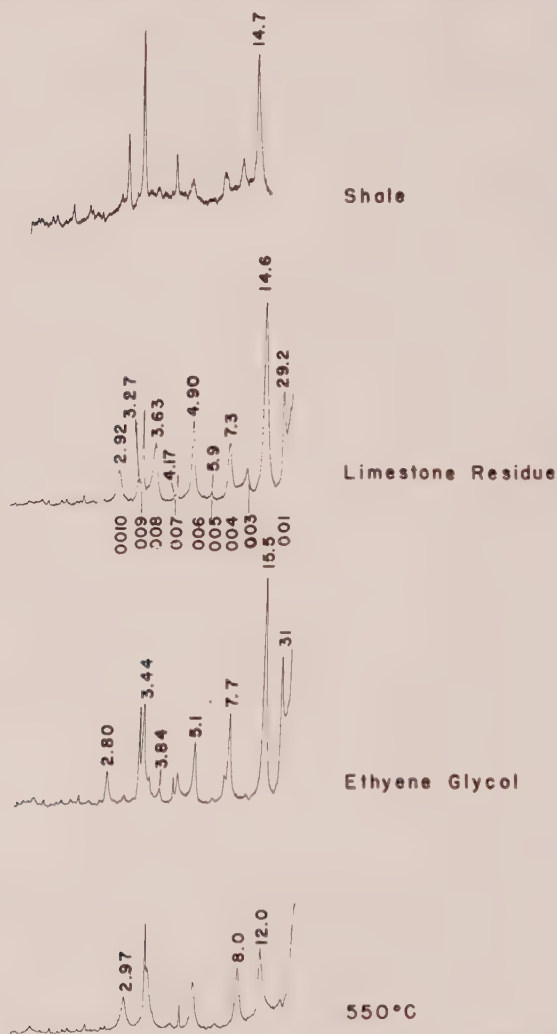


FIG. 11. Regular mixed-layer clays.

layer clay containing 50 per cent 10-Å illite layers and 50 per cent 14-Å montmorillonite layers a given 120-Å interval will contain approximately five 10-Å illite layers and five 14-Å montmorillonite layers. The stronger scatterer will be encountered 5 times per 120 Å, yielding an ill-defined

period of about 24 Å, for which a diffuse scattering maximum is to be observed. In the regular mixed-layer clay the value 120/5 is consistent and the resulting  $x$ -ray reflection is relatively sharp; however, in random mixed-layer clays this value is only a statistical average of a population which may have values ranging from 120/1 to 120/12 and thus the reflection is relatively broad and variable.

Regular mixed-layer clays are not common in sedimentary rocks but several have been found. Figure 11 contains  $x$ -ray diffraction patterns of two identical regular mixed-layer clays. One is from a Pennsylvanian shale and the other from a Mississippian limestone residue. The two clays in Fig. 11 are apparently composed of alternating 14.4-Å chlorite layers and 14.8-Å vermiculite layers. The (001) value is approximately 29.2 Å. Listed below are the calculated (00 $l$ ) series and the measured (00 $l$ ) series for untreated, glycolated, and heated patterns.

	Untreated		Ethylene Glycol		550° C.	
	Calculated (Å)	Measured (Å)	Calculated (Å)	Measured (Å)	Calculated (Å)	Measured (Å)
001	29.2	29	31.0	31	24.0	—
002	14.6	14.6	15.5	15.5	12.0	12.0
003	9.73	9.7	10.3	10.2	8.0	8.0
004	7.30	7.30	7.75	7.7	6.0	6.0
005	5.84	5.90	6.20	6.2	4.8	4.77
006	4.87	4.90	5.16	5.12	4.0	4.0
007	4.17	4.17	4.43	4.40	3.43	3.40
008	3.65	3.63	3.88	3.84	3.0	2.97
009	3.24	3.27	3.44	3.44	2.66	—
0010	2.92	2.92	3.10	3.08	2.40	—

When the sample is treated with ethylene glycol the vermiculite layers are increased to 17 Å and an (001) value of approximately 31 Å is obtained. Again the (00 $l$ ) sequence is integral. The 7.2-Å and 3.60-Å lines indicate the presence of some unmixed chlorite. After being heated to 550° C. the vermiculite layers collapse to 10 Å and the chlorite to 14.0 Å. The resulting (00 $l$ ) values are an integral sequence of 24 Å.

#### EXAMPLE OF HOW MIXED-LAYER CLAY MINERALS CAN BE USED FOR ZONATION

Mixed-layer clays, though presumably quite sensitive to environmental changes, can be found to maintain a consistent ratio through several hundred feet of sedimentary rock. Figure 12 contains  $x$ -ray patterns

which illustrate how it is possible to zone a thick section of sedimentary rock by using the mixed-layer clays.

The sample from the upper zone, in addition to illite, contains a mixed-layer vermiculite-chlorite, with the vermiculite layers predominating.

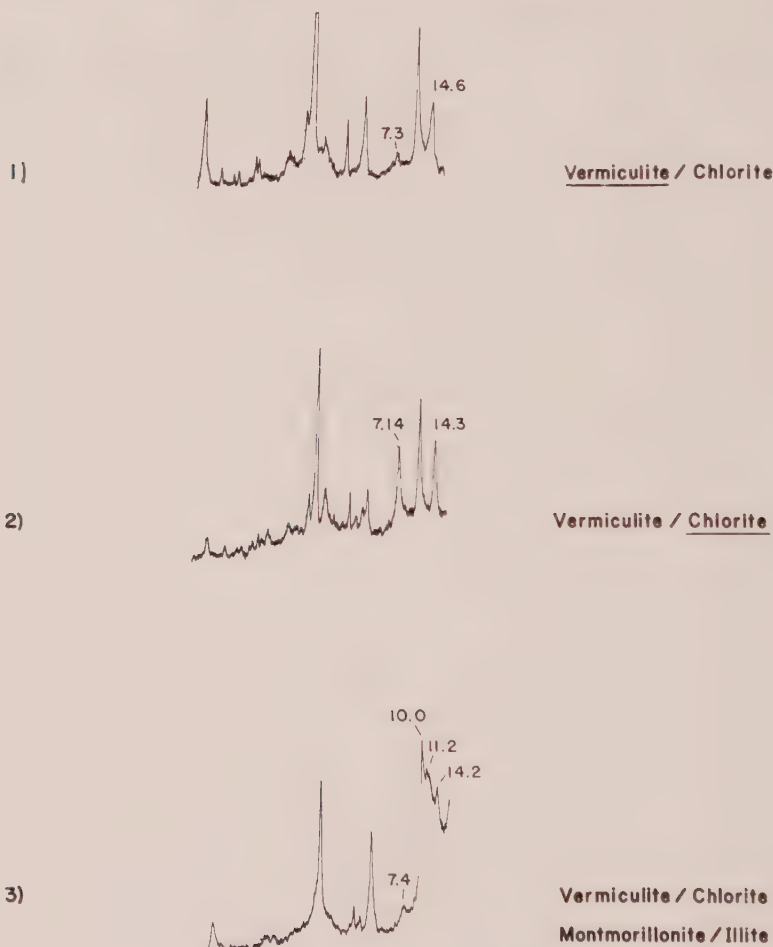


FIG. 12. Shale zonation with mixed-layer clays.

The sample from the middle zone contains a mixed-layer vermiculite-chlorite which is predominantly chlorite. The lower zone contains illite, a mixed-layer illite-montmorillonite, and a mixed-layer vermiculite-chlorite.

## ACKNOWLEDGMENT

The writer wishes to express his gratitude to Dr. W. F. Bradley for the many discussions on mixed-layer clays.

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# QUANTITATIVE DIFFERENTIAL THERMAL ANALYSES OF CLAY AND OTHER MINERALS

H. W. VAN DER MAREL,  
*Agricultural Experiment Station and Institute for  
Soil Research, T.N.O., Groningen, Netherlands.*

## ABSTRACT

The shape and the intensity of the thermal curve of minerals, when analyzed by the *d.t.a.* method, are strongly influenced by amorphous coatings and disordered structures on the surface of the particles (Beilby layer), and furthermore by differences in particle and or crystallite size, the degree of crystallinity of the crystallites and ion substitutions in the crystal structure. Examples are given.

Especially in the group of natural clay minerals, variations in these phenomena are numerous. Consequently an accurate quantitative determination of the amount of a clay mineral in a sample by means of the *d.t.a.* method, is impossible. Thus it was found that even kaolinite from pure well known deposits has a heat of transformation which varies from 100 to 176 and from 23 to 43 cal./g. for its endothermal (ca 600° C.) and exothermal reaction (ca 980° C.) respectively.

For other minerals reliable results can be obtained only if the conditions under which the specimens are formed in nature are practically always the same and the minerals are more-over of pure chemical composition, i.e., circumstances, which are mostly seldom found. These conclusions are based on experiments where errors due to the differential thermal analysis *d.t.a.* apparatus are eliminated by calibration of the instrument with a standard chemical, use of the same sample holder and thermocouple, dilution of the mineral with the inert reference material and packing the mixtures always in the same way in the sample holder.

## INTRODUCTION

Quantitative *d.t.a.* has been applied by several authors as a rapid, inexpensive and accurate method for the determination of the amount of a mineral—see e.g. Norton (1940) for kaolinite, hydrargillite and diasporé in bauxites from Dutch Guiana, Kiyoura and Sata (1950), and Murray et al. (1951) for calcite in limestones. This method of analysis was also used by Vold (1949) to determine the amount of stearic or benzoic acid in a sample from its heat of fusion. The result should be accurate to within a few per cents according to the authors. Grimshaw and Roberts (1953) suggest that the accuracy of the *d.t.a.* method can be increased by diluting the samples 1:3 with inert (calcined) alumina before the test. For in this case the conductivity of the samples investigated should be nearer to that of the inert  $\alpha\text{-Al}_2\text{O}_3$  reference material which is used in a *d.t.a.*

However, Speil et al. (1945) found for undiluted samples of kaolinite from various origins, deviations of about 30% in their heats of transformation. De Bruijn and Van der Marel (1954) who investigated aside from kaolinite also other minerals from various localities, arrived at

deviations of 100 to 200%. These large deviations cannot be ascribed merely to the heat of conductivity being unequal for all the samples investigated. According to the authors, they should be caused by differences in particle size, degree of crystallinity of the crystallites and substitutions in the crystal structure of the samples investigated. Another factor is that loss of heat flowing away from the samples through the various thermocouples cause deviations of ca 30% according to De Josselin de Jong (1955).

It is the purpose of this article to obtain data about the variation in thermal effects of minerals from various origins, and also about the most important factors by which heat of transformation of a certain mineral is affected. All the *d.t.a.* tests described in the experimental part of this article have been performed with the apparatus constructed and described by De Bruijn (1954) and which has been in operation in the Laboratory of Soil Mechanics at Delft (Netherlands) since 1952.<sup>1</sup>

The nickel-block oven contains 6 holes in which 5 samples can be analyzed simultaneously, the remaining hole being reserved for the inert reference material, usually  $\alpha$ - $\text{Al}_2\text{O}_3$  previously heated for several hours at ca. 1300° C. It is provided with Pt/Pt, Rh thermocouples, a Boersma amplifier and a Brown recorder.

## EXPERIMENTS

Factors by which the thermal transformations of a mineral are affected when it is heated and the effect registered by the *d.t.a.* method, are:

### (1) *Particle size of the mineral investigated*

Many authors have found that the finer the particle size of the sample, the lower is the actual temperature at its transformation and the smaller is the peak area—see Van Nieuwenberg (1929), Speil et al. (1945), Laws and Page (1946), Lippman (1952), Martin and Russell (1952) and Robertson et al. (1954) for kaolinite, Caillère and Hénin (1947) for antigorite, Kerr and Kulp (1948) for dickite, Van Schuylenborgh and Arens (1950) and Kulp and Trites (1951) for goethite, Perkins (1950) and Martin and Russell (1952) for montmorillonite, Sabatier (1950) for chlorite, Barshad (1950) for paragonite, Barshad (1950), Mackenzie and Milne (1953), De Bruijn and Van der Marel (1954) for muscovite. De Bruijn and Van der Marel (1954) for pyrophyllite, Faust (1949), Graf

<sup>1</sup> The author is indebted to Prof. Ir. E. C. W. A. Geuze, Director of the Laboratory for Soil Mechanics, Delft, for use of this apparatus and to H. Labrie for assistance in the analyses.

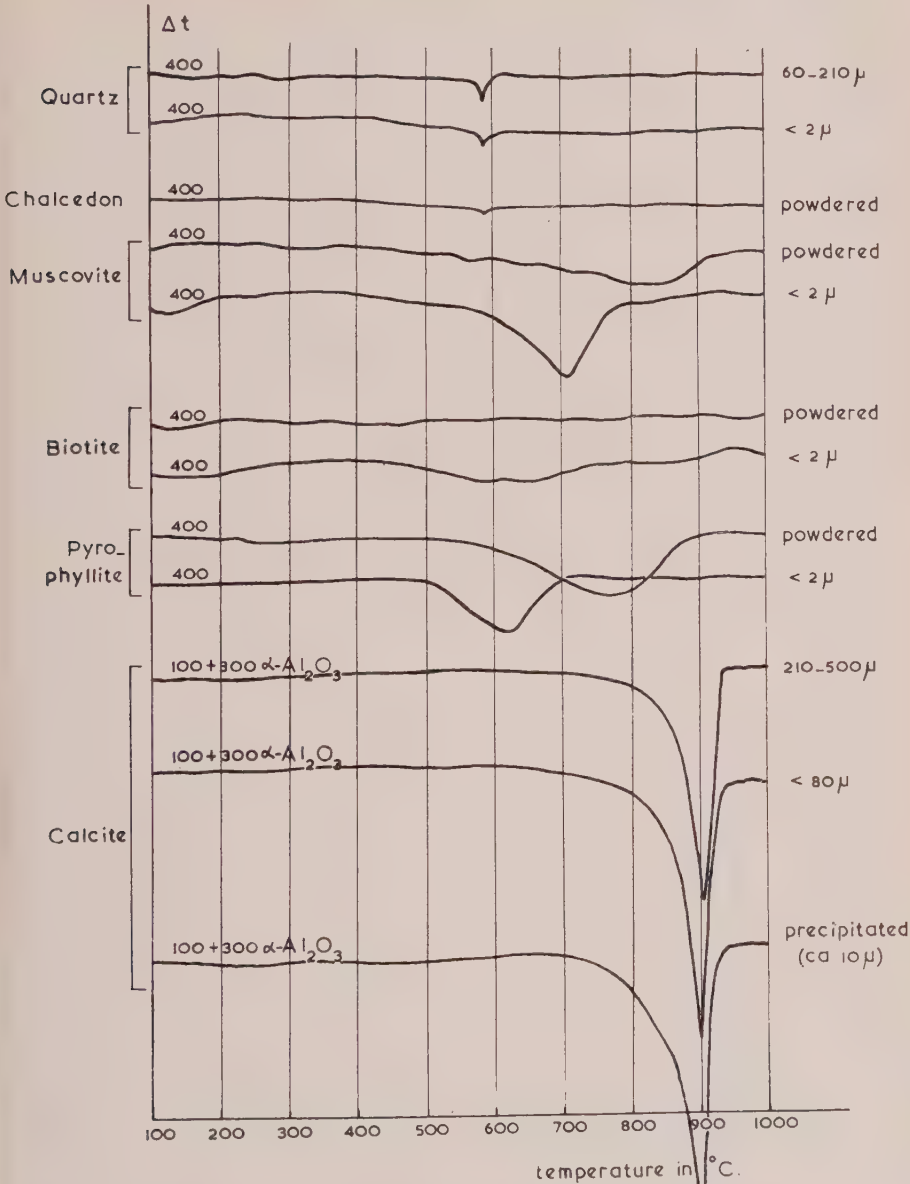
(1952), Bradley et al. (1953) for dolomite, Kulp et al. (1951) for calcite. Also quartz when occurring in nature as small crystallites (chalcedony), which after  $x$ -ray analysis shows a perfect structure, only demonstrates a small negligible  $\alpha/\beta$  *d.t.a.* effect—see Berkelhamer (1944) and Fieldes (1952).

Apparently, the smaller the particle or the crystallite size, the smaller are the forces needed for the transformations inside the crystals. Even a qualitative identification of many minerals may be impossible because by differences in particle size the peak temperature may be shifted about 200° C. to lower temperatures and the intensity of the endothermal effect may be largely decreased—see the examples in Speil et al. (1945) for kaolinite, Caillère and Hénin (1947) for antigorite and De Bruijn and Van der Marel (1954) for muscovite and pyrophyllite. Figure 1 shows examples demonstrating the large effect of particle size on the thermal reactions of quartz, chalcedony, muscovite, biotite, pyrophyllite and calcite.

Another difficulty is that when particle size decreases there is an increased influence of the amorphous or vitreous—so called Beilby (1921)—layer on the thermal effects. This layer has chemical-physical properties quite different from those of the original crystalline material. Thus Laws and Page (1946), Perkins (1950), Parkert et al. (1950), Dragsdorf et al. (1951) found that by grinding kaolinite in a ball mill, the original 600° C. endothermal reaction was almost completely lost and a severe fogging appeared on the  $x$ -ray photo. Moreover, the cation exchange capacity of the material has largely increased. After treatment with 0.1 *N* HCl the residue gave sharp diffraction patterns identical with those of the unground sample. Their conclusion was that by grinding, amorphous permutite-like substances or amphoteric  $\text{Al}(\text{OH})_3$  and  $\text{H}_4\text{SiO}_4$  groups are formed, coating the particles with a thin layer and having a quite different physical-chemical behaviour as compared with the original mineral.

Fieldes (1952), using the silt fraction of a New Zealand soil, which after  $x$ -ray diffraction contained 50% of  $\alpha$ -quartz, found only 5% of it by a *d.t.a.*, the mineral having its usual 573° C. inversion peak. Quantitative analysis of quartz by means of  $x$ -ray and differential thermal analysis and the occurrence of an amorphous layer on the surface of fine quartz particles has been extensively studied, because fine dust in the air of mining districts may contain quartz which causes a lung disease (silicosis) when inhaled continuously.

It was found that the Beilby layer could be removed by treating the sample with a borate buffer—see Clelland et al. (1952)—or by etching with HF—see Clelland and Ritchie (1952), Nagelschmidt et al. (1952).



Note: The curves of quartz, muscovite and pyrophyllite are derived from De Bruijn and Van der Marel (1954) - Part II. pg. 407.

FIG. 1. D.T.A. of various minerals in different grades of fineness.  
Weight of samples analyzed in mg.



Grimshaw and Roberts (1953)—and that the intensity of the original  $\alpha/\beta$  transformation could be restored.<sup>2</sup>

## (2) Degree of crystallinity of the mineral investigated

According to Grimshaw et al. (1945), Grim (1947) and Murray (1954) the endo- and exothermal reaction of the poorly crystallized kaolinites (fire clay, ball clay) are broader, of less intensity and they begin at a lower decomposition temperature than the corresponding reactions of the better crystallized ones. The first is caused by loss of OH from the crystal structure and the latter by the formation of  $\gamma$   $\text{Al}_2\text{O}_3$ .

Caillère and Hénin (1948b) ascribe the difference in peak temperature of the endothermal reaction of dickite, kaolinite and metahalloysite,<sup>3</sup> being ca. 580° C., 550° C. and 525° C., respectively, to differences in the degree of crystallinity. The first is the best crystallized and the last the poorest. Apparently a mineral with a high degree of crystallinity needs more energy for its transformation to a new structure than a mineral of poor crystallinity. According to Speil et al. (1945), Kerr and Kulp (1948), Bramao et al. (1950, 1952) and Glass (1954), metahalloysite (and halloysite) may also be distinguished from kaolinite by the shape of the endothermal reaction, *i.e.* the peak for metahalloysite (halloysite) returns to the base line at a faster rate than it departs, whereas well crystallized kaolinites show about equal rates. The shape ratio of the thermogram, *i.e.*  $\tan \alpha / \tan \beta$ , in which  $\alpha$  = the angle between the perpendicular to the peak and the descending side and  $\beta$  = the corresponding angle on the ascending side, has been proposed by Bramao et al. (1952) as a convenient means to distinguish kaolinite from metahalloysite (halloysite). The reason of this different behaviour of the above minerals is, according to Kerr and Kulp (1948), that the sheets in the metahalloysite (halloysite) structure are superimposed in a less orderly manner than in kaolinite. The occurrence of halloysite (metahalloysite) in nature as small lath-shaped and not as plate-like particles such as kaolinite, is also supposed

<sup>2</sup> The thickness of the Beilby layer on quartz particles was computed by Clelland (1951), Dempster (1951), Clelland et al. (1952), Clelland and Ritchie (1952) and Dempster and Ritchie (1952, 1953) from the decrease in density, the decrease in *d.l.a.* effect and the increase in  $\text{SiO}_2$  dissolved by the borate buffer between the original and the ground kaolinite sample. They found 0.03–0.05  $\mu$ , 0.11–0.15  $\mu$  and 0.02–0.03  $\mu$  respectively. Nagelschmidt et al. (1952), Gibb et al. (1953) found from x-ray and electron diffraction analyses 0.03  $\mu$  and 0.03–0.06  $\mu$ , respectively. Meldau and Robertson (1952) arrived at 0.03 to 0.20  $\mu$  for augite, hornblende, fluorite and aragonite by electron diffraction analysis.

<sup>3</sup> In this paper the terms halloysite and metahalloysite, the latter resulting from the former by heating at ca. 45° C., have been used to designate the phases  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 4\text{H}_2\text{O}$  and  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$ , respectively, also called endellite and halloysite, respectively by other investigators.

to be caused by stresses in the crystal resulting from this particular structure.

By *x*-ray analysis the poorer degree of crystallinity of halloysite and metahalloysite as compared with kaolinite can easily be observed on the photo—see e.g. van der Marel (1950). Therefore, due to a lack of perfection of the individual crystallites, rather than to their smaller size, the *d.t.a.* curve of halloysite (metahalloysite) is not symmetrical as in kaolinite, but asymmetrical. However, de Bruijn and van der Marel (1954) and Robertson et al. (1954) observed many well crystallized kaolinites, verified by *x*-ray analysis, to have an asymmetric endothermal reaction. In Plate I are some examples. Thus the size of the crystallites and amorphous coatings—see under (1)—on the particles have also affected the shape and the intensity of the endothermal effect of kaolinite.

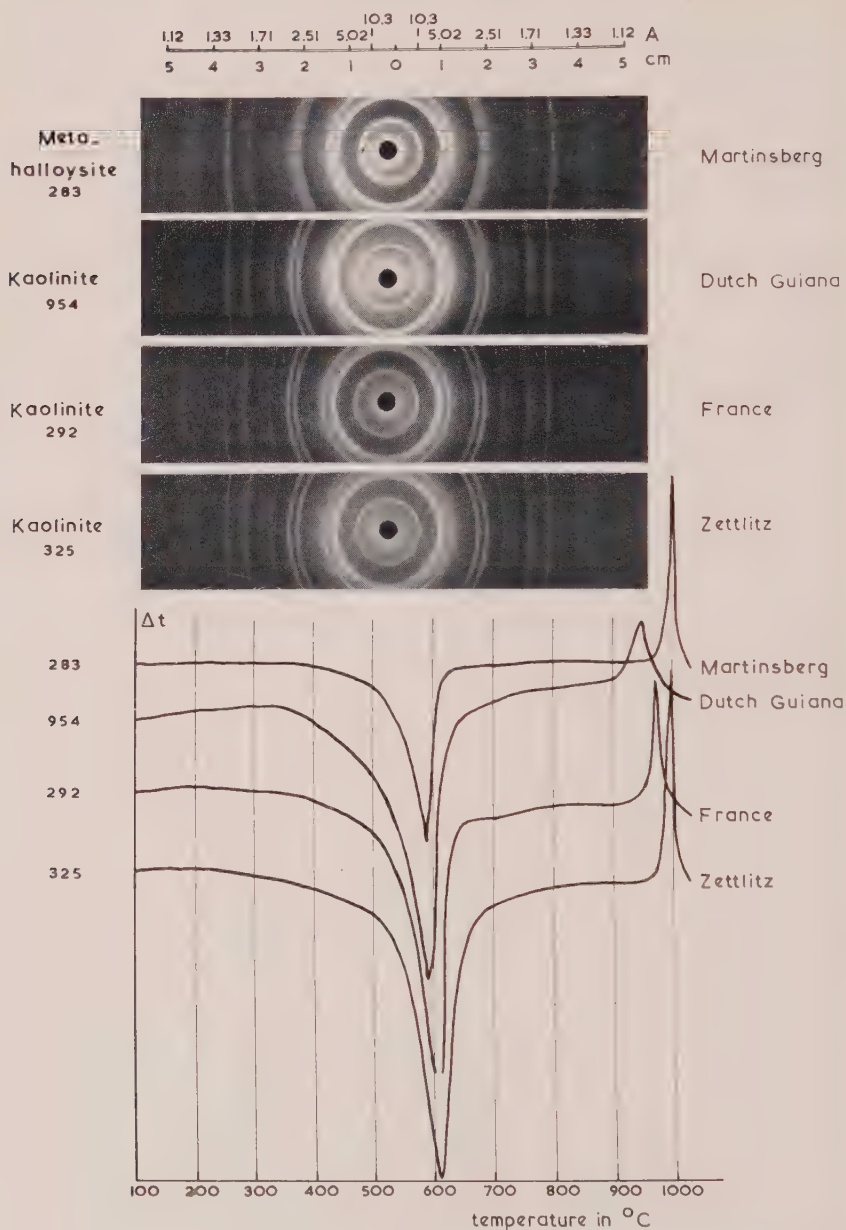
### (3) *Ion substitutions in the crystal structure of the mineral investigated*

According to Orcel (1935) and afterwards Kelley and Page (1943), Caillère and Hénin (1947, 1948*a,b*, 1949), Kulp et al. (1951), Graf (1952) the introduction of iron into the structure of a mineral, substituting aluminum, magnesium or silicon, shifts the endothermal peak temperature to lower grades and changes its intensity. According to Caillère and Hénin (1947, 1948*a,b*, 1949) the kind of binding of OH in the crystal structure of a mineral is related to its decomposition temperature. In this way OH of talc, antigorite and brucite is bound with decreasing strength in the same order as here mentioned as they have endothermal effects at 950° C., 650° C. and 400° C., respectively.

Page (1943) and Kelley and Page (1943) found that when Al is the predominant constituent of the octahedral sheet of the 2:1 minerals, the water is given off at a lower temperature than when Mg is the predominant constituent. Grim and Rowland (1942) found that iron-free Texas montmorillonite does not show its exothermal peak (caused by the formation of a spinel) until at ca. 1050° C. As a contrast magnesium-rich Otay montmorillonite does not show a distinct exothermal reaction because enstatite is formed at ca. 950° C. instead of spinel—see Earley et al. (1953). In Fig. 2 is demonstrated the evident effect of ion substitutions on the shape and the intensity of the *d.t.a.* curve for some montmorillonites, kaolinites and carbonates—see also the examples given by other investigators.

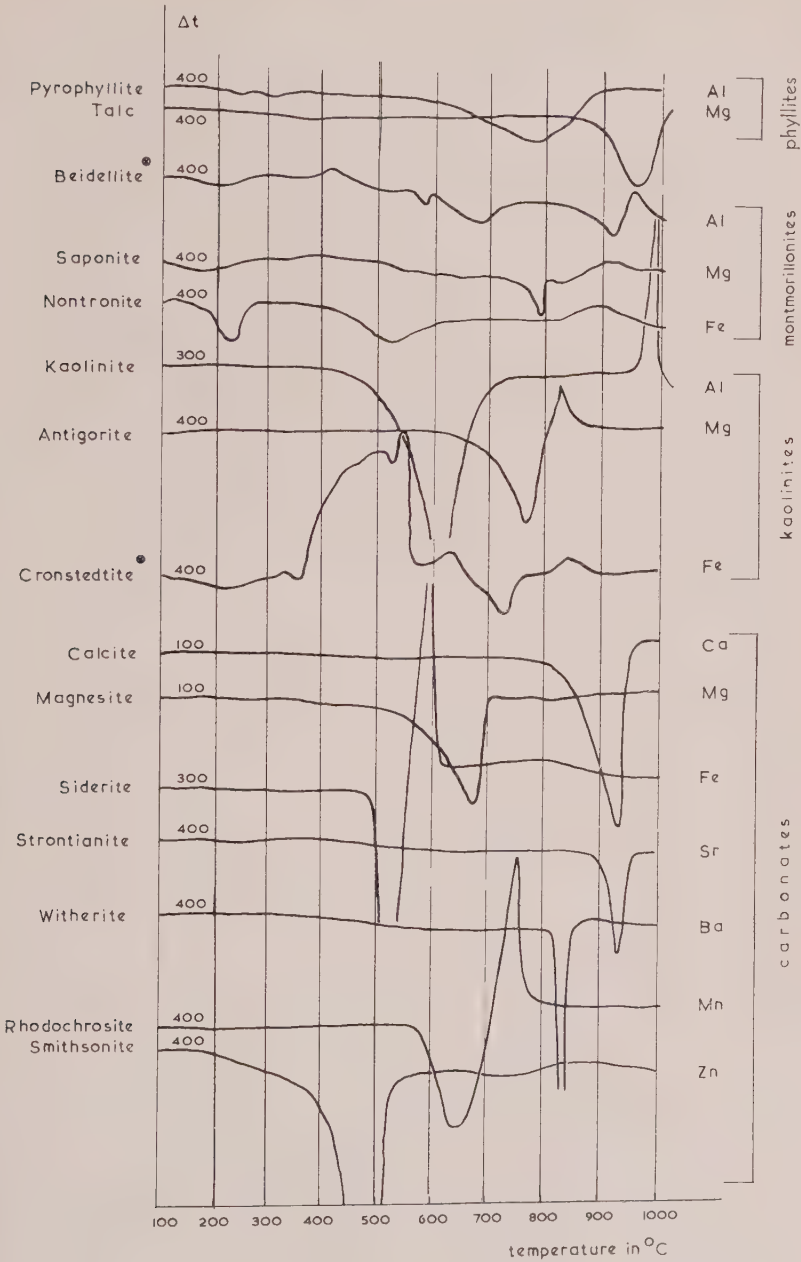
### (4) *Different exchangeable cations in the mineral investigated*

Hendricks et al. (1940), Caillère and Hénin (1944), Barshad (1948, 1950) and Arens (1951) found that the kind of exchangeable cation affects the shape and the intensity of the low temperature endothermal



Note: all the samples were dried 1 hour at 105° C. before the *d.t.a.*

PLATE I. X-ray and *d.t.a.* analyses of metahalloysite and kaolinite  $<2\mu$  with an asymmetric shape of their endothermal reaction at 600° C. Furthermore of kaolinite  $<2\mu$  from Zettlitz (Czechoslov.) with a symmetric shape as usual.



⊗ The author is indebted for these valuable samples to Dr. S. B. Hendricks, U. S. Department of Agriculture, Beltsville, Maryland, (U.S.A.).

FIG. 2. D.T.A. of pyrophyllite and talc and some montmorillonites, kaolinites and carbonates. Weight of samples analyzed in mg.



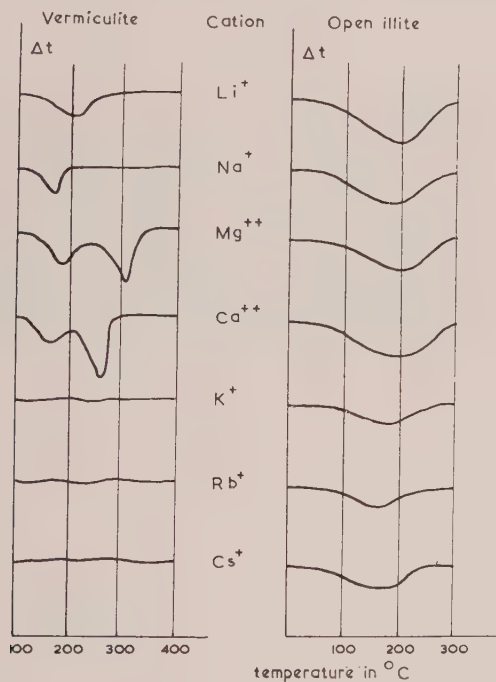
reaction of montmorillonite. Faust (1951) arrived at the same result for saucornite, *e.g.* a mineral representing the Zn-end member of the montmorillonite series.<sup>4</sup> The low endothermal reaction of montmorillonite is caused by loss of planar water, broken bound water and water bound to adsorbed cations. The latter is the most firmly held and thus has an endothermal reaction at higher temperature than the foregoing.

A very large effect on the *d.t.a.* diagram of saturating a mineral with different cations, has been observed for vermiculite by Barshad (1948), Walker and Milne (1950) and Arens (1951). This mineral contracts its plates when saturated with  $K^+$ ,  $NH_4^+$ ,  $Rb^+$  and  $Cs^+$  ions thereby expelling the water molecules between the layers, when saturated with  $Ca^{++}$ ,  $Mg^{++}$ , or  $Sr^{++}$  ions—see Gruner (1939), Barshad (1948, 1950), Walker (1950). Mackenzie (1950) has attempted to correlate the peak temperatures as found by Hendricks et al. (1940) for montmorillonite saturated with various cations, with the hydration energies of these ions according to Bernal and Fowler (1933). A correlation was found, except for lithium. However Greene-Kelley (1953) pointed out that the activation energy detected by the *d.t.a.* method is not directly related to the hydration energy of the cation concerned. It will also depend on the charge density between the layers. In this way cations which give double peaks for vermiculite, may give single peaks for montmorillonite. Of course minerals with a small cation exchange capacity such as kaolinite—see Arens (1951)—will not manifest significantly the phenomenon of its *d.t.a.* curve being influenced by the kind of adsorbed cation. In Fig. 3 are some examples demonstrating the effect of the kind of adsorbed cations on the low temperature thermal reaction of vermiculite and open illite (ammersooite). The latter mineral also contracts its plates when treated with  $K^+$ ,  $NH_4^+$ ,  $Rb^+$  and  $Cs^+$  ions thereby expelling its cation bound  $H_2O$  molecules from between the layers—see Van der Marel (1954). In order to eliminate the disturbing effect of the weaker bound  $H_2O$  molecules, the samples were dried (in this case 1 hour at  $105^\circ C.$ ) before they were analyzed by the *d.t.a.* method. Moreover the samples were analyzed under conditions where errors due to the apparatus could not disturb the results. As pointed out by De Josselin de Jong (1956) these errors are: dilution of the sample with the inert reference material ( $\alpha-Al_2O_3$ ), packing the samples always in the same way in the sample holder, covering the thermocouples with sufficient material so that the heat flow in the neighbourhood of the thermocouples always follows the same geometrical (cylindrical) pattern. Finally the sample holder + its

<sup>4</sup> By montmorillonite is meant here, as in common usage, the high aluminum member of the montmorillonite series with some slight replacement of  $Al^{3+}$  by  $Mg^{2+}$  and no replacement of  $Si^{4+}$  by  $Al^{3+}$ .

thermocouple was calibrated with a standard chemical (in this case  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ ) before and after each mineral/ $\alpha\text{-Al}_2\text{O}_3$  mixture was analyzed.

As may be observed from the results, the effect of the kind of exchangeable cation is smaller for the open illite as for vermiculite. However, the total cation exchange capacity of the open illite is only ca. 65 m.e./100



Note: All samples dried 1 hour before the *d.t.a.* and diluted 3:1 with  $\alpha\text{-Al}_2\text{O}_3$ .

FIG. 3. *D.t.a.* of vermiculite and open illite (ammersooite) saturated with different cations.

g. as against ca. 120 m.e./100 g. for vermiculite. Therefore, also the total amount of  $\text{H}_2\text{O}$  molecules which influence the thermal effect of open illite is smaller than that of vermiculite.

##### (5) Chemical reactions between minerals in the sample investigated

Decomposition of pure dolomite,  $\text{CaMg}(\text{CO}_3)_2$  takes place in two steps. First the magnesite component loses its  $\text{CO}_2$  at ca.  $800^\circ\text{C}$ . and thereafter the calcite component loses its  $\text{CO}_2$  at ca.  $900^\circ\text{C}$ .—see Flood (1950). However, Stutterheim et al. (1951), Heady (1952) and Webb

(1953) found that in an intimate mixture of dolomite and  $\text{Ca(OH)}_2$ , the normal large endothermal reaction due to the decomposition of  $\text{MgCO}_3$  in the dolomite, was hardly noticeable. This effect was caused by the instantaneous, exothermal reaction between the  $\text{CO}_2$  evolved from the  $\text{MgCO}_3$  fraction of the dolomite with the  $\text{CaO}$  resulting from the prior decomposition of the  $\text{Ca(OH)}_2$ . In consequence the first endothermal effect of dolomite was almost completely cancelled, but the second endothermal effect of dolomite was enlarged as its effect was increased by the endothermal reaction of the formed  $\text{CaCO}_3$  having the same peak temperature as the  $\text{CaCO}_3$  component of dolomite. On the other hand Webb (1953) found for molar mixtures of  $\text{MgCO}_3$  and  $\text{CaO}$  or  $\text{Ca(OH)}_2$ , that a  $\text{MgCO}_3$  peak was obtained which was followed by only a small  $\text{CaCO}_3$  peak. Thus the same reaction took place as in the foregoing example but only less readily. In mixtures of quartz and calcite, the endothermal reaction of calcite is hindered by an exothermal reaction of calcium silicate, following immediately upon the former—see Lippmann (1952).

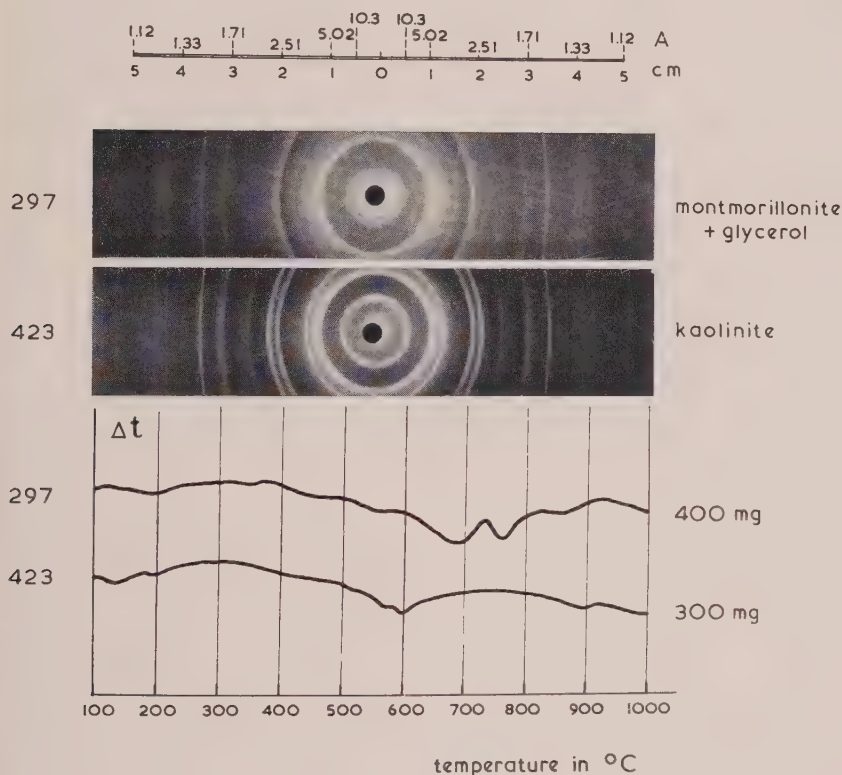
(6) *Other disturbing components in the sample investigated*

According to Grimshaw et al. (1945), Heady (1952) and De Bruijn and Van der Marel (1954) exothermal effects caused by small amounts of organic matter (humus) are often so pronounced as to reduce, or even to eliminate any endothermal effect which may take place simultaneously. By treating the sample before the *d.t.a.* with  $\text{H}_2\text{O}_2$ , slightly decomposable organic material may be oxidized. Many soils and shales, however, contain organic matter which cannot be destroyed with  $\text{H}_2\text{O}_2$ , e.g. cellulose, lignin and elemental carbon. According to Mackenzie and Lakin (1953) graphitic (crystallized) carbon is less easily oxidized in air than amorphous carbon and thus shows an oxidation peak at a much higher temperature.

It should be mentioned here that Allaway (1949) could magnify the endothermal effect of montmorillonite, beidellite and nontronite by investigating samples saturated with piperidine. The latter is first reduced to carbon and then yields hydrogen with the water vapour escaping from the crystal at its decomposition temperature and which has a great heat of combustion when reacting with the oxygen of the air. However, for kaolinite having only a small base exchange capacity, or for samples mixed with finely divided carbon, this method has proved to be valueless. Evidently, a mechanical mixture of inert clay with piperidine or carbon burns in a way much different from a clay which contains adsorbed piperidine cations.

If organic matter is present, the only way to avoid errors is to analyze the sample in vacuum or argon atmosphere—see Rowland and Lewis

(1951). A number of workers—see Budnikov and Bobrovink (1938), Berg (1943, 1945), Schwob (1950) and Graf (1952)—have described the evident, decreasing effect of only small amounts of soluble alkali and alkaline earth chlorides and carbonates, upon the thermal decomposition of dolomite. Caillère and Hénin (1948*b*) observed for mixtures of NaCl



Note: the samples were dried 1 hour at 105° C. before the *d.t.a.*

PLATE II. X-ray and *d.t.a.* analysis of montmorillonite  $<2\mu$  and kaolinite  $<2\mu$  which contain  $\text{Na}_2\text{CO}_3$ .

and KCl that the endothermal peak temperature—being for each component in its pure state ca. 800° C.—is even shifted to 660° C. in the 50% mixture. Moreover, they found for NaCl, KCl,  $\text{CaCO}_3$ ,  $\text{CaSO}_4$  mixtures that the endothermal reaction of calcite at 900° C. may disappear completely and that new ones are produced. This is due to various reactions occurring during the heating of the sample. Gruver et al. (1949) found a strong suppression of the thermal reactions of kaolinite when 5% NaCl



or 5%  $\text{Na}_2\text{CO}_3$  was added to the sample and that the suppression of the exothermal reaction was decreased more than that of the endothermal reaction.

Plate II shows the results of *x-ray* and *d.t.a.* analyses of kaolinite and montmorillonite which have been purified by sedimentation in Atterberg cylinders, 0.01 N NaOH being used as peptisator. As with these fine clays the NaOH could not be washed off entirely, the dried samples which were analyzed, still contained (ca 4%)  $\text{Na}_2\text{CO}_3$  (through absorption of  $\text{CO}_2$  from the air). It may be concluded that the thermal curves of these samples, which by *x-ray* analysis are pure well crystallized kaolinite and montmorillonite, have evidently been changed by the impurity.

#### (7) *Volume changes in the sample investigated*

Shrinking, sintering or melting of the sample during heating, affect its thermal properties and therefore also the shape and the intensity of the *d.t.a.* curve—see Norton (1939), Berg (1945) and De Josselin de Jong (1956). In Fig. 4 are some striking examples—see also the examples given by other investigators. Thus magnesite, calcite, brucite, hydrargillite lose ca. 52%, 44%, 30% and 30% respectively of their weight at the decomposition temperature. As a result of the decreased conductivity, after the decomposition of these minerals, the base line comes at a higher level than before. This effect may be decreased by mixing the samples with  $\alpha\text{-Al}_2\text{O}_3$ . Zinnwaldite and lepidolite melt at their decomposition temperature = ca. 900° C. Thereby the thermocouple is covered with a tight glassy substance and the contact between the sample and the sample holder is disturbed. As a result of the decreased conductivity of the sample, the *d.t.a.* curve moves downwards, thus making any quantitative analysis impossible. Another inconvenience is that after the test the thermocouple has become useless for further analysis. Incomplete oxidation of carbo lignin during heating caused by lack of sufficient oxygen, is manifested by a gradually downward movement of the *d.t.a.* curve. In this case only reliable results for the heat of combustion of this organic—see Fig. 4—can be obtained if the sample is diluted 1:29 with  $\alpha\text{-Al}_2\text{O}_3$ .

From the above it may be concluded that with a *d.t.a.* great differences may be expected in the intensity, the area and the shape of the thermal peaks of minerals from different origins. This holds especially for clay minerals where variations in particle size of the crystallites and chemical composition (ion substitutions) in the tetrahedral and octahedral layers, may be numerous in nature—see f.e. the analyses of Ross and Hendricks (1954) and Early et al. (1953)—and the minerals being moreover, mostly coated with various kinds of amorphous substances which also influence the thermal reactions.

Figure 5 shows the *d.t.a.* results of  $<2\mu$  samples of montmorillonite<sup>5</sup> from various, pure deposits. They were obtained by sedimentation of the original samples in Atterberg cylinders with 0.05 N  $\text{NH}_4\text{OH}$  as peptisa-

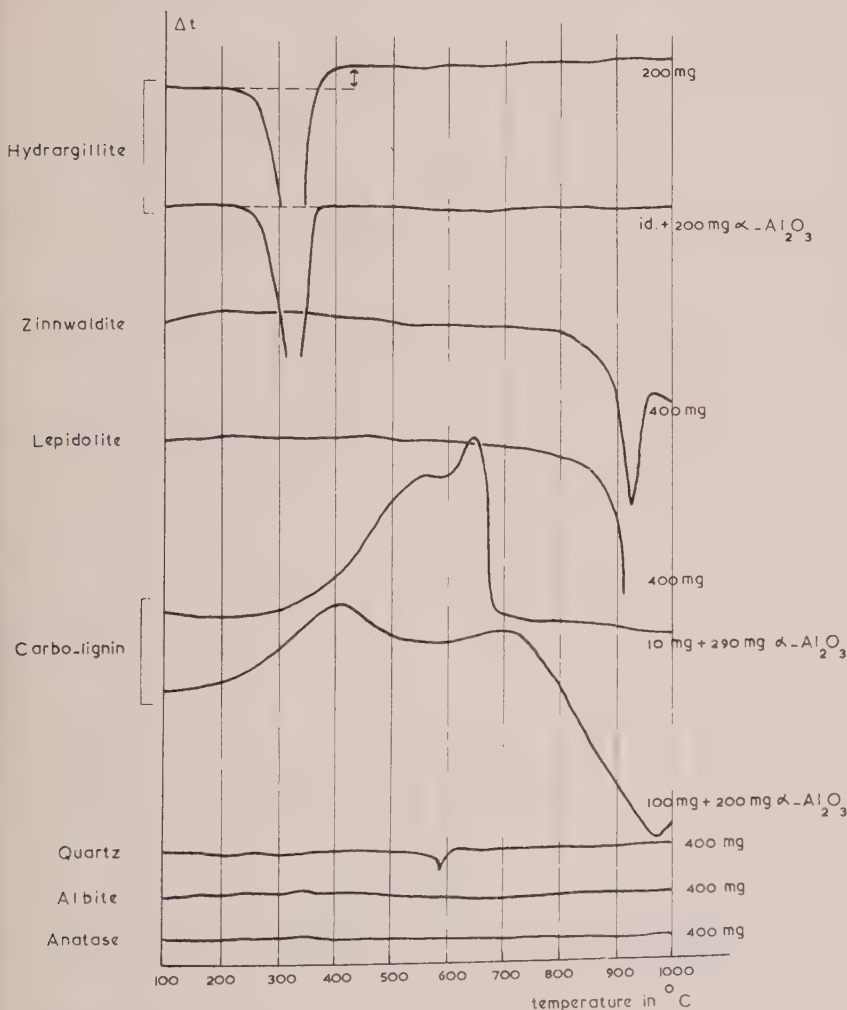


FIG. 4. *D.t.a.* of minerals which lose large amounts of water (hydrargillite), which melt (zinnwaldite, lepidolite) or which burn off (carbo-lignin) when heated. Furthermore of quartz and of some inert minerals (albite, anatase).

tor. The excess of electrolytes was washed out by filtration and the purity of the dried samples was checked by *x*-ray diffraction after the glycerol method of MacEwan (1946). The curves show evidently that the peak

<sup>5</sup> See for this qualification note 4.

temperature, the intensity and even the relative intensities between the high temperature endothermal and exothermal reaction of this clay mineral vary considerably when it is registered by the *d.t.a.* method.

Figure 6 shows the *d.t.a.* results of the  $<2\mu$  separate of soils of various

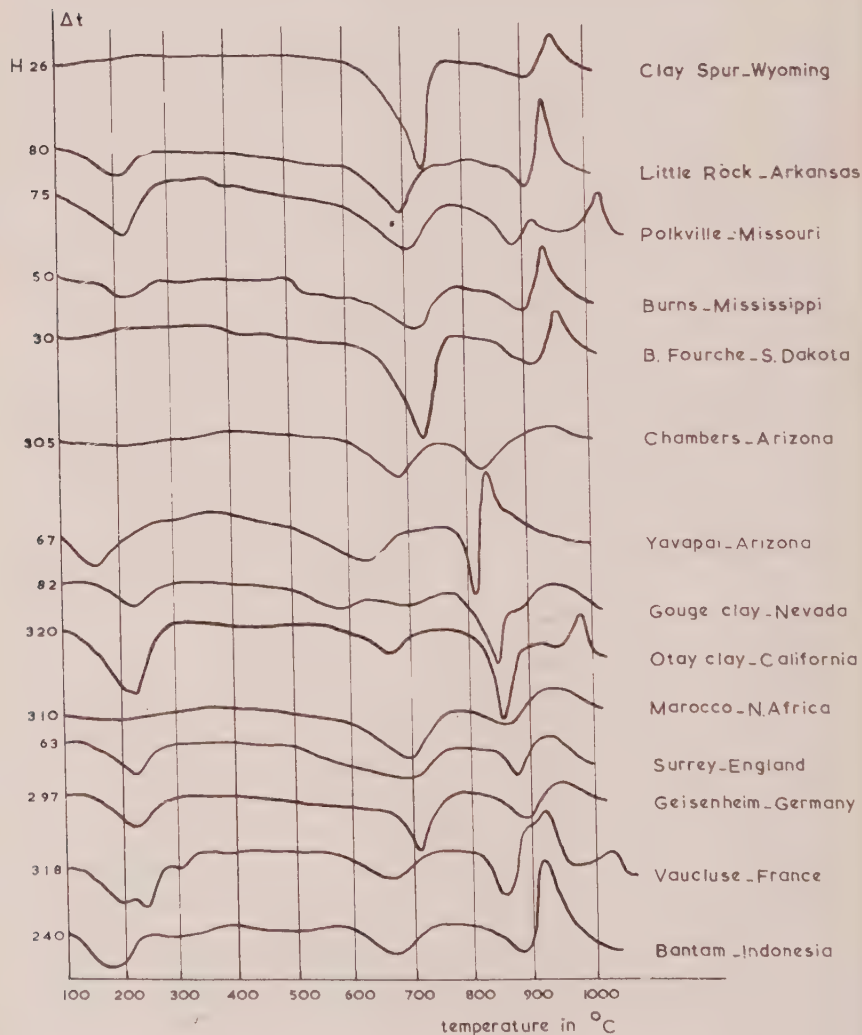
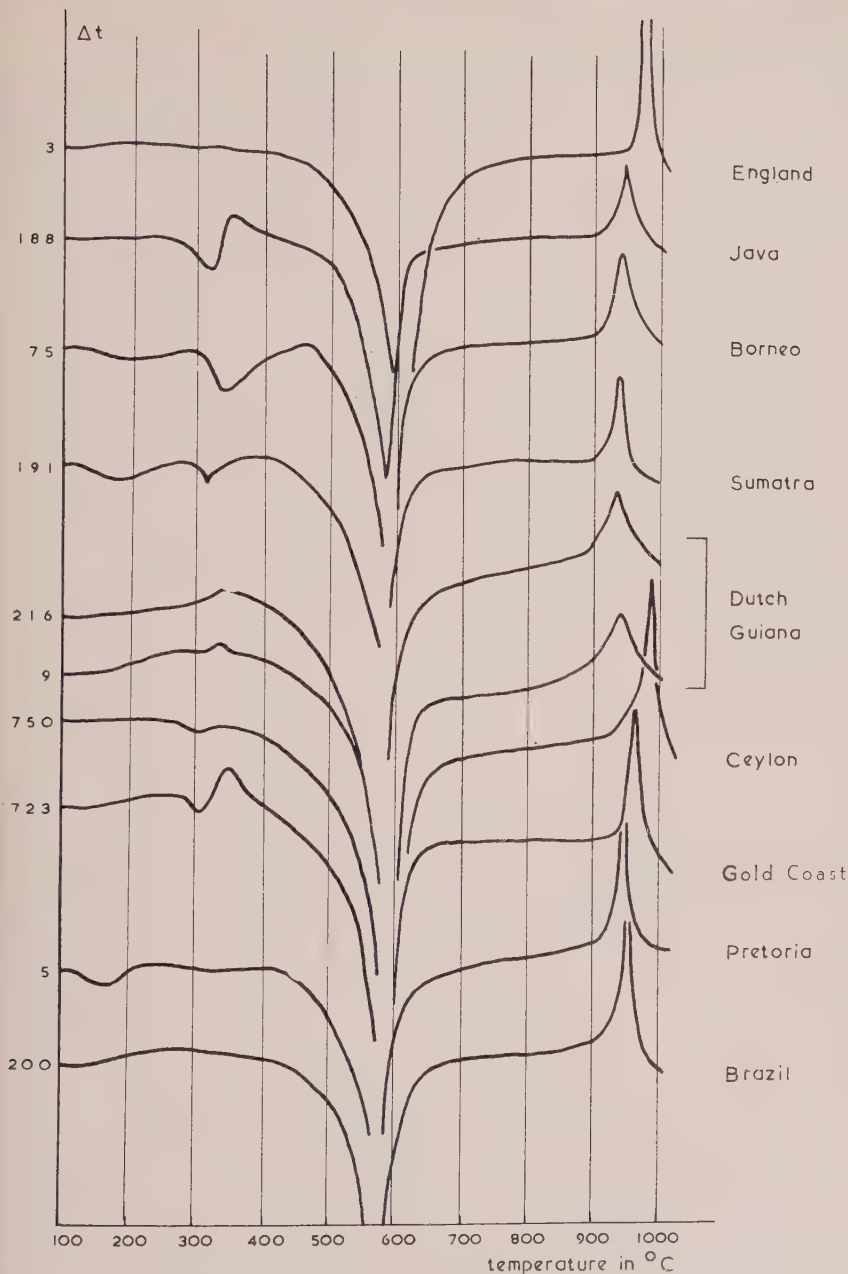


Fig. 5. *D.t.a.* of dried (1 hour at  $105^{\circ}\text{C}.$ ) samples  $<2\mu$ , each 400 mg. of pure (verified by  $x$ -ray analysis) montmorillonite from different localities.



Note: Endothermal effect at ca 325 $^{\circ}$  C. and exothermal effect at ca 350 $^{\circ}$  C. of some samples caused by small amounts of hydrargillite and iron oxides respectively.

FIG. 6. *D.t.a.* of dried (1 hour at 105 $^{\circ}$  C.) samples  $<2\mu$ , each 300 mg. of pure white kaolinite from Cornwall (England). Furthermore of high graded kaolinites  $<2\mu$  from soils from different localities.

origin which after x-ray analysis contain practically only kaolinite. The separate was obtained by first treating the original samples with  $\text{H}_2\text{O}_2$  (to destroy easily decomposable organic matter). They were afterwards treated in the same way in Atterberg cylinders, etc., as is described for montmorillonite. The *d.t.a.* curves show that also in this case, the peak temperature, the intensity and the relative peak areas of the endothermal

TABLE 1. PEAK AREA OF PURE, WHITE, WELL CRYSTALLIZED (VERIFIED BY  $\alpha$ -RAY ANALYSIS) DRIED (1 HOUR AT  $105^\circ\text{C.}$ ) KAOLINITE  $<2\mu$  AND OF DRIED (1 HOUR AT  $105^\circ\text{C.}$ ) POWDERED  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$  (STANDARD) DILUTED WITH  $\alpha\text{-Al}_2\text{O}_3$  1.50:2.50 EQUALLY PACKED IN THE SAMPLE HOLDER AND ANALYZED WITH THE SAME THERMOCOUPLE. *D.t.a.* OF KAOLINITES ALTERNATING WITH THOSE OF STANDARD  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$  SAMPLES

No.	Origin	KAOLINITE			STANDARD
		Endothermal (ca $600^\circ\text{C.}$ ) in $\text{cm}^2$	Exothermal ca( $950^\circ\text{C.}$ ) in $\text{cm}^2$	Endothermal Exothermal	Endothermal (ca $150^\circ\text{C.}$ ) in $\text{cm}^2$
					31.8
39)	Dutch Guiana	19.8	4.6	4.30	33.3
954)		14.6	3.4	4.29	32.8
1220	Gabon, Congo	22.0	4.8	4.58	32.2
163	Cornwall, England	26.6	5.6	4.75	32.6
325	Zettlitz, Tsechoslov	19.5	4.7	4.15	32.0
40	Provence	22.2	4.8	4.62	31.2
292	Reims	17.2	4.0	4.30	32.6
460	Java	18.8	4.4	4.27	33.4
14	Bangka	22.3	5.3	4.21	31.8
653	Pem. Siantar	21.8	4.4	4.95	32.6
128	Tandj. Balei	20.9	3.9	5.13	31.5
202	Dhong Thuan, Thailand	17.2	3.5	4.91	33.2
326	Murfreesboro, Ark.	24.7	6.0	4.12	31.7
327	Bath, Carol.	26.5	6.3	4.21	30.3
329	Macon, Georgia	22.1	6.0	3.68	30.7
330	Dry Branch, Georgia	24.0	5.5	4.36	33.5
115	New Jersey	22.7	4.7	4.83	31.1
328	Mesa Alta, Nw. Mexico	25.7	5.5	4.67	30.5
					32.5
	Arithmetical mean ( $\bar{x}$ )				32.06

Note: By calculation was found for the  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O} \rightarrow \text{CuSO}_4 \cdot \text{H}_2\text{O}$  transformation at ca  $150^\circ\text{C.}$ :

Standard error of the mean ( $s_{\bar{x}}$ ) =  $0.22\text{ cm}^2 = 0.69\%$ .

Standard error of the single determination ( $s$ ) =  $0.97\text{ cm}^2 = 3.02\%$ .

and the exothermal reaction vary considerably.

To determine the magnitude of the variations in the heat of transformation of kaolinite, being the least variable clay mineral, *d.t.a.* were performed on the  $<2\mu$  separates of pure, white, well crystallized (verified by x-ray analysis) kaolinite and under conditions where errors due the *d.t.a.* method are eliminated.

The results are summarized in Table 1. It may be concluded that under these optimum conditions, the measured heat of transformation of the



standard chemical used, is practically constant. It has a standard deviation of the single determination (*s*) of only 3.02% for its first ( $\text{CuSO}_4 \cdot 5\text{H}_2\text{O} \rightarrow \text{CuSO}_4 \cdot \text{H}_2\text{O}$ ) endothermal effect (peak temperature = ca. 150° C.). However, the endothermal and the exothermal reaction of kaolinites from different origin has a peak area which varies from 14.6 cm.<sup>2</sup> to 25.7 cm.<sup>2</sup> and from 3.4 cm.<sup>2</sup> to 6.3 cm.<sup>2</sup> respectively.<sup>6</sup> The relative intensities of the two thermal reactions varies from 3.7 to 5.1. Another difficulty is that many minerals have reactions which overlap each other for a large part e.g. kaolinite and quartz, kaolinite and illite, glauconite and illite—see for further samples also Table 1 of De Bruijn and Van der Marel (1954), Part II, page 412. Only exceptionally the overlapping can be eliminated by use of a slow-heating rate, or by heating the sample in vacuum-, in nitrogen- or in carbon dioxide atmosphere—see for the latter Rowland and Lewis (1951) and Rowland and Beck (1952) in case of dolomite.

Then there is still the difficulty that varieties of a mineral may occur in nature, which are of the same structure and composition, but differing largely in their thermal effect—see for  $\alpha$  quartz McDowall and Dunn (1947), McDowall and Vose (1947, 1952), Fieldes (1952), Keith and Tuttle (1952) and Lewcock and Wylde (1953).

### DISCUSSION

Particle size, degree of crystallinity of the crystallites, kind and amount of ion substitutions and amorphous coatings (Beilby layer) may vary considerably for a certain mineral of different origin. As they greatly influence the heat of transformation of a mineral as registered by a *d.t.a.* the application of this method for quantitative purposes will therefore be very restricted. This conclusion holds especially for the group of clay minerals. The same factors have so far resisted also a quantitative determination of this group of minerals by the *x*-ray method.

The quantitative *d.t.a.* method can only be applied to well crystallized minerals of well defined chemical composition if they give sharp diagrams of great intensity and if they are formed in nature under practically the same conditions. However, attention should also be paid in this case to the sample investigated, as very fine crystallites and small amounts of chlo-

<sup>6</sup> For the decomposition of 150 mg.  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$  to  $\text{CuSO}_4 \cdot \text{H}_2\text{O}$  at ca 150° C. 33 calories are needed (see tables of constants). This thermal effect is registered by the *d.t.a.* apparatus here used as a peak area of 32.06 cm.<sup>2</sup> (standard deviation of the mean = 0.22 cm.<sup>2</sup>). Thus the variation in the thermal transformations of pure kaolinite when heated in a *d.t.a.* oven, is as follows:

endothermal reaction 100 to 176 cal. per gram  
exothermal reaction 23 to 43 cal. per gram

rides or carbonates of alkali and alkaline earths may largely disturb the thermal effects. However, there lies an open and at present only accidentally explored field for the *d.t.a.* method in the study of the heat of transformation of  $H_2O$  and other polar molecules bound to rest valences at the surface of minerals, or to the exchangeable cations of these minerals. Another possibility for the *d.t.a.* method is the study of ion substitutions in a certain mineral and their place in the crystal structure in combination with chemical and x-ray analysis. In these cases small differences in the physical behaviour of the  $H_2O$  molecules (e.g. when stirred to an ice-like state) or in the strength by which OH ions are bound in a crystal, may cause large differences in the peak area or the peak temperature of a *d.t.a.* Their registration can be made more accurate and therefore the application of the *d.t.a.* technique as a very sensitive calorimetric method for quantitative purposes enlarged, (1) by carrying out the analyses at a high heating rate in vacuum-, argon- or carbon dioxide atmosphere; (2) by use of the more sensitive Au, Pd/Pt, Rh (Pallaplat) thermocouple which is also resistant to oxidation at the higher oven temperatures;<sup>7</sup> (3) by placing the thermocouple not inside the sample holder as usual, but as recently suggested by Boersma (1955), outside it. In that case the peak area is no more dependent on the heat conductivity, the heat capacity or volume changes of the mineral investigated, but merely on the produced reaction heat of the mineral sample and a certain calibration constant of the apparatus.<sup>8</sup>

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<sup>7</sup> Manufactured by W. C. Heraeus, G.M.B.H., Hanau (Germany). It has a E.M.F. at 100° C., 300° C., 600° C. and 900° C. of 2.86, 10.60, 24.72 and 40.35 mV, respectively. The corresponding thermal effects of a Pt/Pt, Rh thermocouple are 0.64, 2.31, 5.23 and 8.45, respectively.

<sup>8</sup> See for details on the factors affecting the application of quantitative mineral analysis with the commonly used *d.t.a.* equipments and the construction and the perspectives of the Boersma sample holder, the articles by De Josselin de Jong (1956) and Boersma (1955).

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# A NEW EQUATION RELATING INDEX OF REFRACTION AND SPECIFIC GRAVITY\*

ROBERT D. ALLEN, *U. S. Geological Survey, Claremont, Calif.*

## ABSTRACT

A new equation relating index of refraction and specific gravity is proposed:  $\alpha\beta\gamma/d=k$ . The terms  $\alpha\beta\gamma$  and  $k$  are referred to as refractive capacity and specific refractive capacity, respectively. The new equation is compared with 3 classical equations by means of 12 groups of polymorphous compounds. Specific refractive capacities are in closer agreement for  $\text{Al}_2\text{O}_3$ ,  $\text{SiO}_2$ ,  $\text{CaCO}_3$ ,  $\text{Al}_2\text{SiO}_5$ , and  $\text{NaAlSiO}_4$  than are the classical constants. All equations yield satisfactory agreement between modifications of  $\text{MgSiO}_3$ ,  $\text{CaSiO}_3$ ,  $\text{ZnS}$ , and  $\text{Na}_2\text{SO}_4$ . Specific refractive capacities for the respective polymorphs of  $\text{TiO}_2$ ,  $\text{As}_2\text{O}_3$ , and  $\text{Sb}_2\text{O}_3$  show gross discrepancies, considerably greater than those found with the older equations. Such discrepancies demonstrate that refraction of light is a constitutive as well as an additive property and are thought to result from variations in manner of aggregation.

Specific refractive capacities for the common oxides are tabulated. Average index of refraction is calculated for 26 compositions by means of (1)  $\alpha\beta\gamma/d=k$  and (2)  $(n-1)/d=k$ ; in the majority of these examples, the former expression yields indices closer to those experimentally measured.

## INTRODUCTION

During the past century, three principal equations relating index of refraction to specific gravity have been proposed. The existing formulas (Larsen and Berman, 1934) are listed in Table 1. The Lorentz and Lorenz

TABLE 1. EQUATIONS RELATING INDEX OF REFRACTION AND SPECIFIC GRAVITY

Author(s)	Equation	Designation of Constant
Gladstone and Dale	$\frac{n-1}{d}=k$	Specific refractivity
Lorentz and Lorenz	$\frac{n^2-1}{n^2+2} \cdot \frac{1}{d}=k$	Specific refraction
Lichtenecker	$\frac{\log n}{d}=k$	_____

equation is based on the electromagnetic theory of light, whereas the others are of empirical origin.

Although numerous articles dealing with relationships among chemical

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composition, index of refraction, and density in glass have recently appeared, at present only approximate generalized formulas for wide ranges and accurate expressions for limited ranges of composition are possible (Sun, Safford, and Silverman, 1940).

The postulate upon which these relationships are based may be stated as follows: for a definite chemical composition there exists a constant  $k$ , which may be calculated from the index of refraction and density of any one of the composition's physical modifications (solid, liquid, gas; polymorphs). In other words, it is assumed that refraction of light is an additive property dependent solely on concentration and composition. Despite the limited validity of the postulate, this type of equation is useful for computations within restricted ranges where index of refraction is an additive property. Conversely, definite discrepancies may supply useful information about the nature of bonding.

#### BASIS FOR PROPOSED EQUATION

The concept of the indicatrix has been fundamental in redefining refractivity, the capacity of a substance to refract light. The indicatrix (Fletcher, 1892), a geometrical abstraction utilized in optical mineralogy to depict the behavior of light in crystals, may be defined as a three-dimensional geometric figure so constructed that the three principal indices of refraction of light waves in their directions of vibration are equal to its three mutually perpendicular semiaxes. Thus, in isotropic substances, which have but one index of refraction, the indicatrix is a sphere. In uniaxial crystals, which have two indices of refraction, the indicatrix is either a prolate or oblate spheroid of revolution depending on whether the substance is optically positive or negative, respectively. For biaxial crystals, with three indices, the indicatrix takes the general form of a triaxial ellipsoid. In essence, then, the indicatrix is a vectorial representation of the refraction of light in three dimensions.

The refractivity of a substance is here considered to be proportional to the volume of its indicatrix. The formulas expressing volume for the three classes of optical indicatrix are as follows:

Sphere	$4/3\pi n^3$
Spheroid of revolution	$4/3\pi\omega^2\epsilon$
Triaxial ellipsoid	$4/3\pi\alpha\beta\gamma$

In the general case, therefore, refractivity is proportional to the product  $\alpha\beta\gamma$ . The product of the three indices of refraction of a substance  $-\alpha\beta\gamma$  for biaxial crystals,  $\omega^2\epsilon$  for uniaxial crystals, and  $n^3$  for isotropic substances may be defined as refractive capacity. A new synthetic constant,  $k$ , may be obtained from the formula  $\alpha\beta\gamma/d = k$ , where  $d$  is the specific gravity. This value  $k$  may be termed the specific refractive capacity.

TABULATION OF CONSTANTS OF SOME COMMON  
PHYSICAL MODIFICATIONS

The four equations are compared by computation of their respective constants for 12 groups of polymorphous or dimorphous compounds. Data for water-ice and for the isomers of chlorotoluene are also given. Tridymite is omitted from the table because of uncertainty in optical data. For uniaxial and biaxial crystals the mean index of refraction,  $n$ , equals  $\sqrt[3]{\omega^2\epsilon}$  or  $\sqrt[3]{\alpha\beta\gamma}$ ; this quantity must be determined for solution of the classical equations. Specific gravities are calculated (Schlecht, 1944) for crystal modifications of  $\text{SiO}_2$  and  $\text{CaCO}_3$  as well as for compounds which show gross lack of agreement between values of  $k$  when experimental specific gravities are employed. Sources of experimental data are indicated by numbers in parentheses which refer to the bibliography.

It should be pointed out that impurities in minerals may affect the calculations in some cases. For instance, minor amounts of iron, titanium, or chromium may be present in corundum. Columbium, tantalum, and iron have been reported in rutile; iron is usually present in anatase and brookite. It is considered likely, however, that the optical and specific gravity data cited would be essentially correct for the corresponding ideally pure compounds.

## DISCUSSION

Specific refractive capacities are in closer agreement than are the constants calculated by means of the classical equations for polymorphs of the following compositions:  $\text{Al}_2\text{O}_3$ ,  $\text{SiO}_2$ ,  $\text{CaCO}_3$ ,  $\text{Al}_2\text{SiO}_5$ , and  $\text{NaAlSiO}_4$ . Each of these groups is characterized by appreciable differences in specific gravity and indices of refraction. In contrast, for those groups of substances whose polymorphs do not have widely different physical properties, such as,  $\text{MgSiO}_3$ ,  $\text{CaSiO}_3$ ,  $\text{ZnS}$ , and  $\text{Na}_2\text{SO}_4$ , all four equations yield comparable results.

The specific refractive capacities for  $\text{TiO}_2$ ,  $\text{As}_2\text{O}_3$ ,  $\text{Sb}_2\text{O}_3$ , and  $\text{H}_2\text{O}$  show gross discrepancies between the respective modifications of each composition. The differences are considerably greater than those arrived at through use of the older formulas. The immediate explanation lies in the mathematical nature of the various equations. In a series of polymorphous compounds whose indices of refraction are dependent upon variables other than chemical composition and specific gravity, the ratio  $\alpha\beta\gamma/d$  would be subject to greater variations than the corresponding ratios of the previously introduced equations.

It is clear that index of refraction is a constitutive property as well as an additive property, that is, the manner of aggregation of chemical elements is a determining factor besides the kinds and concentrations of

TABLE 2. CONSTANTS OF SOME COMMON PHYSICAL MODIFICATIONS

Formula	Physical modification; crystal system	Specific gravity	Indices of refraction	$\frac{n-1}{d}$	$\frac{n^2-1}{n^2+2} \cdot \frac{1}{d}$	$\frac{\log n}{d}$	$\frac{\alpha\beta\gamma}{d}$
Al <sub>2</sub> O <sub>3</sub> (6, 11)	Corundum; hexagonal (synthetic)	3.98 (calc.)	$\omega$ , 1.768 $\epsilon$ , 1.760	0.192	0.104	0.0620	1.38
	"Third phase"; isometric	3.47	1.696	0.201	0.111	0.0661	1.41
SiO <sub>2</sub> (5, 12, 13)	Low quartz; hexagonal	2.66	$\omega$ , 1.544	0.206	0.119	0.0712	1.39
		2.664 (calc.)	$\epsilon$ , 1.553	0.205	0.119	0.0711	1.39
	Low cristobalite; tetragonal	2.33	1.486	0.209	0.123	0.0738	1.41
		2.342 (calc.)	(mean)	0.208	0.123	0.0735	1.40
TiO <sub>2</sub> (6)	Rutile; tetragonal	4.23	$\omega$ , 2.612	0.403	0.160	0.102	4.68
		4.26 (calc.)	$\epsilon$ , 2.899	0.400	0.159	0.101	4.64
	Brookite; orthorhombic	4.14	$\alpha$ , 2.583	0.392	0.160	0.101	4.35
		4.12 (calc.)	$\beta$ , 2.584 $\gamma$ , 2.700	0.394	0.161	0.102	4.37
	Anatase; tetragonal	3.90	$\omega$ , 2.561	0.394	0.165	0.104	4.18
		4.04 (calc.)	$\epsilon$ , 2.488	0.380	0.159	0.100	4.04
As <sub>2</sub> O <sub>3</sub> (6)	Arsenolite; isometric	3.87	1.755	0.195	0.106	0.0631	1.40
		3.88 (calc.)		0.195	0.106	0.0630	1.39
	Claudetite; monoclinic	4.15	$\alpha$ , 1.871	0.225	0.115	0.0690	1.74
		4.26 (calc.)	$\beta$ , 1.92 $\gamma$ , 2.01	0.219	0.112	0.0672	1.70
Sb <sub>2</sub> O <sub>3</sub> (6)	Senarmontite; pseudoisometric	5.50	2.087	0.198	0.0960	0.0581	1.65
		5.56 (calc.)		0.196	0.0950	0.0575	1.64
	Valentinite	5.76 (exp. and calc.)	$\alpha$ , 2.18 $\beta$ , 2.35 $\gamma$ , 2.35	0.224	0.1017	0.0625	2.09
CaCO <sub>3</sub> (7)	Calcite; hexagonal	2.710	$\omega$ , 1.6585	0.221	0.126	0.0752	1.51
		2.743 (calc.)	$\epsilon$ , 1.4865	0.218	0.125	0.0743	1.49
	Aragonite; orthorhombic	2.95	$\alpha$ , 1.530	0.214	0.121	0.0721	1.47
		2.944 (calc.)	$\beta$ , 1.681 $\gamma$ , 1.685	0.214	0.121	0.0720	1.47
	Vaterite; hexagonal	2.645	$\omega$ , 1.550	0.220	0.126	0.0752	1.49
		(calc.)	$\epsilon$ , 1.645				
MgSiO <sub>3</sub> (11)	Enstatite; orthorhombic	3.175	$\alpha$ , 1.650 $\beta$ , 1.653 $\gamma$ , 1.658	0.206	0.115	0.0688	1.42



TABLE 2 (continued)

Formula	Physical modification; crystal system	Specific gravity	Indices of refraction	$\frac{n-1}{d}$	$\frac{n^2-1}{n^2+2} \cdot \frac{1}{d}$	$\frac{\log n}{d}$	$\frac{\alpha\beta\gamma}{d}$
CaSiO <sub>3</sub> (11)	Clinoenstatite; monoclinic	3.19	$\alpha$ , 1.651 $\beta$ , 1.654 $\gamma$ , 1.660	0.205	0.115	0.0686	1.42
	Wollastonite; monoclinic	2.915	$\alpha$ , 1.616 $\beta$ , 1.629 $\gamma$ , 1.631	0.215	0.121	0.0724	1.47
	Pseudo- wollastonite; monoclinic	2.905	$\alpha$ , 1.610 $\beta$ , 1.610 $\gamma$ , 1.654	0.215	0.122	0.0725	1.48
Al <sub>2</sub> SiO <sub>5</sub> (5)	Kyanite; triclinic	3.60	$\alpha$ , 1.712 $\beta$ , 1.720 $\gamma$ , 1.728	0.200	0.110	0.0654	1.41
	Sillimanite; orthorhombic	3.23	$\alpha$ , 1.659 $\beta$ , 1.660 $\gamma$ , 1.680	0.206	0.115	0.0687	1.43
	Andalusite; orthorhombic	3.15	$\alpha$ , 1.634 $\beta$ , 1.639 $\gamma$ , 1.643	0.203	0.114	0.0681	1.40
NaAlSi <sub>3</sub> O <sub>8</sub> (11)	Carnegieite; triclinic	2.51	$\alpha$ , 1.509 $\beta$ , 1.514 $\gamma$ , 1.514	0.204	0.120	0.0716	1.38
ZnS (6)	Nephelite; hexagonal	2.619	$\omega$ , 1.537 $\epsilon$ , 1.533	0.205	0.119	0.0711	1.38
	Sphalerite; isometric	4.00	2.369	0.343	0.151	0.0936	3.32
	Wurtzite; hexagonal	3.98	$\omega$ , 2.356 $\epsilon$ , 2.378	0.343	0.151	0.0938	3.32
Na <sub>2</sub> SO <sub>4</sub> (7, 11)	Thenardite; orthorhombic	2.664	$\alpha$ , 1.471 $\beta$ , 1.477 $\gamma$ , 1.484	0.179	0.106	0.0636	1.21
H <sub>2</sub> O (3, 6)	Na <sub>2</sub> SO <sub>4</sub> —III; orthorhombic	2.696	1.4825 (mean)	0.179	0.106	0.0634	1.21
	Ice; hexagonal	0.917 0.921 (calc.)	$\omega$ , 1.309 $\epsilon$ , 1.310	0.337 0.336	0.209 0.208	0.128 0.127	2.45 2.44
	Water	1.00	1.335	0.335	0.207	0.125	2.38
ClC <sub>6</sub> H <sub>4</sub> CH <sub>3</sub> (3)	O-Chlorotoluene	1.0817	1.5238	0.484	0.283	0.169	3.27
	M-Chlorotoluene	1.0722	1.5214	0.486	0.284	0.170	3.28
	P-Chlorotoluene	1.0697	1.5199	0.486	0.284	0.170	3.28

elements. No formula, therefore, which involves only index of refraction and density as variables can be expected to produce a constant for all phases and polymorphs of a given chemical composition.

The gross differences in specific refractive capacities of  $\text{TiO}_2$ ,  $\text{As}_2\text{O}_3$ , and  $\text{Sb}_2\text{O}_3$  may be a reflection of variation in bond type from one polymorph to another. Pauling (1948) has pointed out differences in bonding between the respective polymorphs of  $\text{As}_2\text{O}_3$  and  $\text{Sb}_2\text{O}_3$ . Arsenolite ( $\text{As}_2\text{O}_3$ , isometric) and senarmontite ( $\text{Sb}_2\text{O}_3$ , pseudo-isometric), whose specific refractive capacities are relatively low, consist of small molecules of the type  $\text{A}_4\text{X}_6$ . On the other hand, claudetite ( $\text{As}_2\text{O}_3$ , monoclinic) and valentinite ( $\text{Sb}_2\text{O}_3$ , orthorhombic), whose specific refractive capacities are relatively high, have infinite molecules. A somewhat parallel situation is presented by water with discrete molecules (specific refractive capacity = 2.38) and ice with infinite molecules (specific refractive capacity = 2.44). It may be noted that anomalous values of molar refraction\* (Glasstone, 1946, p. 528) have been useful in the determination of bond type in organic compounds. Variations in bond mechanism could account for anomalous values of  $k$  encountered in the modifications of  $\text{TiO}_2$ .

It might also be significant that Ti, As, and Sb are all elements capable of existing in more than one state of positive valence.

#### SPECIFIC REFRACTIVE CAPACITIES OF THE OXIDES

Specific refractive capacities of the oxides have been calculated (1) from the oxides themselves or (2) from compounds containing the oxides. The specific refractive capacity,  $k$ , of a compound of the type  $m\text{AX} \cdot n\text{BX}$ , where  $m$  is the weight percentage of  $\text{AX}$  and  $n$  is the weight percentage of  $\text{BX}$ , is given by the equation

$$\frac{mk_{\text{AX}} + nk_{\text{BX}}}{100} = K_{\text{AX} \cdot \text{BX}}.$$

If  $K_{\text{AX} \cdot \text{BX}}$  and either  $k_{\text{AX}}$  or  $k_{\text{BX}}$  are known, the equation may be solved for the unknown  $k$  value. Many of the specific refractive capacities tabulated below have been calculated in this manner. Of course, this computation presupposes additivity of  $k$  values.

References in Table 3 are indicated in parentheses.

#### CALCULATION OF AVERAGE INDEX OF REFRACTION FOR MINERALS, ARTIFICIAL COMPOUNDS, AND GLASSES

Average index of refraction may be calculated for a compound whose specific refractive capacity and specific gravity are known. (Conversely,

\* Molar refraction is defined as  $(n^2 - 1)/(n^2 + 2) \cdot M/d$ , where  $M$  is the molecular weight.

density may be calculated if average index of refraction and specific refractive capacity are known.) Specific refractive capacity is given by the earlier described equation

$$\frac{mk_{AX} + nk_{BX}}{100} = K_{AX \cdot BX}.$$

Calculated average indices of refraction for minerals, artificial compounds, and glasses, obtained from (1)  $\alpha\beta\gamma/d=k$  and (2)  $(n-1)/d=k$ , are tabulated in Table 4. Experimental indices and data used in the calculations are included. Values of  $k$  for the Gladstone and Dale equation were taken from Larsen and Berman (1934). Chemical analyses are recalculated to 100 per cent. As before, references are indicated in parentheses.

In 15 of 26 compositions the average indices calculated by means of the new equation are substantially nearer to the experimental values than are those obtained from the Gladstone and Dale equation. In nine compositions the two equations yield calculated indices which differ from the experimental by similar increments. In two compositions indices calculated by the Gladstone and Dale equation are significantly nearer to the experimental. It must be pointed out that certain oxides, such as  $H_2O$ ,  $CO_2$ ,  $Fe_2O_3$ ,  $TiO_2$ , are not characterized by constant specific refractive capacities for all compounds (see Table 3), and calculations involving these oxides should be carried out with caution.

#### SUMMARY

A new equation relating index of refraction and specific gravity is proposed:  $\alpha\beta\gamma/d=k$ . The term  $\alpha\beta\gamma$  is referred to as refractive capacity and is proportional to the volume of the indicatrix;  $k$  is designated as specific refractive capacity.

The new equation is compared with three earlier equations by computation of their respective constants for 12 groups of polymorphous compounds. Specific refractive capacities for the respective polymorphs of  $Al_2O_3$ ,  $SiO_2$ ,  $CaCO_3$ ,  $Al_2SiO_5$ , and  $NaAlSiO_4$  are in closer agreement than the classical constants; each of these groups is characterized by appreciable differences in specific gravity and indices of refraction. In contrast, every equation yields satisfactory agreement between modifications of  $MgSiO_3$ ,  $CaSiO_3$ ,  $ZnS$ , and  $Na_2SO_4$ ; each of these groups is characterized by small differences in specific gravity and indices of refraction.

The specific refractive capacities for  $TiO_2$ ,  $As_2O_3$ ,  $Sb_2O_3$ , and  $H_2O$  show large discrepancies, greater than the constants calculated from the older formulas, between the respective modifications of each composition. These anomalies are attributed to variations in the manner of aggregation of atoms.

Specific refractive capacities for the common oxides are tabulated.

TABLE 3. SPECIFIC REFRACTIVE CAPACITIES OF THE COMMON OXIDES

Oxide	Specific refractive capacity	Source of data
Ag <sub>2</sub> O	1.07	AgNO <sub>3</sub> (11)
Al <sub>2</sub> O <sub>3</sub>	1.38	Al <sub>2</sub> O <sub>3</sub> (11)
As <sub>2</sub> O <sub>3</sub>	1.40	Arsenolite (6)
	1.74	Claudetite (6)'
B <sub>2</sub> O <sub>3</sub>	1.60	CaB <sub>2</sub> O <sub>4</sub> (11)
	1.65	Mg <sub>3</sub> B <sub>2</sub> O <sub>6</sub> (11)
	1.70	B <sub>2</sub> O <sub>3</sub> glass (3)
BaO	0.909	Celsian (5)
BeO	1.71	BeO (11)
Bi <sub>2</sub> O <sub>3</sub>	0.820	Bi <sub>2</sub> O <sub>3</sub> (3)
CO <sub>2</sub>	1.31	Calcite-aragonite (7)
	1.40	Dolomite (7)
CaO	1.63	Anorthite (5)
CrO <sub>3</sub>	2.36	K <sub>2</sub> CrO <sub>4</sub> (11)
Cr <sub>2</sub> O <sub>3</sub>	2.09	FeCr <sub>2</sub> O <sub>4</sub> (12)
	3.01	Cr <sub>2</sub> O <sub>3</sub> (11)
Cs <sub>2</sub> O	0.834	Cs <sub>2</sub> SO <sub>4</sub> (11)
Cu <sub>2</sub> O	3.31	Cu <sub>2</sub> O (11)
CuO	1.75	CuSO <sub>4</sub> (11)
FeO	1.42	FeAl <sub>2</sub> O <sub>4</sub> (12)
	1.54	Fayalite (5)
Fe <sub>2</sub> O <sub>3</sub>	2.49	Andradite (5)
	5.80	Hematite (6)
H <sub>2</sub> O	2.25	Gypsum (7)
	2.38	Water (3)
	2.44	Ice (6)
HgO	1.41	HgO (3)

TABLE 3—(continued)

Oxide	Specific refractive capacity	Source of data	
K <sub>2</sub> O	1.33	Orthoclase	(5)
Li <sub>2</sub> O	2.20	Li <sub>2</sub> O	(11)
MgO	1.46	MgO	(11)
MnO	1.44	Tephroite	(5)
N <sub>2</sub> O <sub>5</sub>	1.60	BaN <sub>2</sub> O <sub>6</sub>	(11)
Na <sub>2</sub> O	1.28	Albite	(5)
P <sub>2</sub> O <sub>5</sub>	1.03	Na <sub>4</sub> P <sub>2</sub> O <sub>7</sub>	(3)
	1.26	KH <sub>2</sub> PO <sub>4</sub>	(11)
PbO	1.08	Pb(NO <sub>3</sub> ) <sub>2</sub>	(3)
Rb <sub>2</sub> O	0.885	Rb <sub>2</sub> SO <sub>4</sub>	(11)
SO <sub>3</sub>	1.14	Anhydrite	(7)
Sb <sub>2</sub> O <sub>3</sub>	1.65	Senarmontite	(6)
	2.09	Valentinite	(6)
SiO <sub>2</sub>	1.39	Quartz	(5)
SnO <sub>2</sub>	1.20	Cassiterite	(3)
SrO	1.04	Celestite	(7)
	1.11	SrAl <sub>2</sub> Si <sub>2</sub> O <sub>8</sub>	(11)
ThO <sub>2</sub>	1.03	ThO <sub>2</sub>	(3)
TiO <sub>2</sub>	4.18	Anatase	(6)
	4.35	Brookite	(6)
	4.68	Rutile	(6)
WO <sub>3</sub>	1.05	CaWO <sub>4</sub>	(11)
ZnO	1.16	ZnAl <sub>2</sub> O <sub>4</sub>	(11)
	1.45	Zincite	(6)
ZrO <sub>2</sub>	1.73	Zircon	(5)



TABLE 4. CALCULATED AVERAGE INDICES OF REFRACTION FOR MINERALS, ARTIFICIAL COMPOUNDS, AND GLASSES

Substance	Composition	<i>k</i> values employed		Specific gravity	Average index of refraction		
		$\alpha\beta\gamma/d$	$(n-1)/d$		Experi- mental	Calculated	
						$\alpha\beta\gamma/d$	$(n-1)/d$
Tridymite (5)	SiO <sub>2</sub>	1.39	0.207	2.30	1.471	1.473	1.476
Baddeleyite (5)	ZrO <sub>2</sub>	1.73	0.201	5.7±	2.17	2.144	2.146
Spinel (5)	MgO · Al <sub>2</sub> O <sub>3</sub>						
	MgO 28.34%	1.46	0.200	3.60	1.718	1.716	1.702
	Al <sub>2</sub> O <sub>3</sub> 71.66%	1.38	0.193				
	<i>K<sub>S</sub></i>	1.403	0.1950				
Mullite (5)	3Al <sub>2</sub> O <sub>3</sub> · 2SiO <sub>2</sub>						
	Al <sub>2</sub> O <sub>3</sub> 71.80%	1.38	0.193	3.23	1.644	1.646	1.636
	SiO <sub>2</sub> 28.20%	1.39	0.207				
	<i>K<sub>M</sub></i>	1.382	0.1969				
Rhodonite (3)	MnO · SiO <sub>2</sub>						
	MnO 54.15%	1.44	0.191	3.72	1.739	1.740	1.738
	SiO <sub>2</sub> 45.85%	1.39	0.207				
	<i>K<sub>R</sub></i>	1.417	0.1983				
Pyrope (5)	3MgO · Al <sub>2</sub> O <sub>3</sub> · 3SiO <sub>2</sub>						
	MgO 30.01%	1.46	0.200	3.51	1.705	1.704	1.707
	Al <sub>2</sub> O <sub>3</sub> 25.29%	1.38	0.193				
	SiO <sub>2</sub> 44.70%	1.39	0.207				
	<i>K<sub>P</sub></i>	1.409	0.2013				
Grossularite (5)	3CaO · Al <sub>2</sub> O <sub>3</sub> · 3SiO <sub>2</sub>						
	CaO 37.35%	1.63	0.225	3.53	1.735	1.734	1.743
	Al <sub>2</sub> O <sub>3</sub> 22.64%	1.38	0.193				
	SiO <sub>2</sub> 40.01%	1.39	0.207				
	<i>K<sub>G</sub></i>	1.477	0.2106				
Anorthite (5)	CaO · Al <sub>2</sub> O <sub>3</sub> · 2SiO <sub>2</sub>						
	CaO 20.16%	1.63	0.225	2.765	1.583	1.583	1.589
	Al <sub>2</sub> O <sub>3</sub> 36.65%	1.38	0.214				
	SiO <sub>2</sub> 43.19%	1.39	0.207				
	<i>K<sub>A</sub></i>	1.435	0.2065				
Iron barium silicate (11)	FeO · BaO · 4SiO <sub>2</sub>						
	FeO 15.44%	1.54	0.187	3.33	1.620	1.611	1.591
	BaO 32.95%	0.909	0.127				
	SiO <sub>2</sub> 51.61%	1.39	0.207				
	<i>K</i>	1.255	0.1776				
Thenardite (5)	Na <sub>2</sub> SO <sub>4</sub>						
	Na <sub>2</sub> O 43.64%	1.28	0.181	2.69	1.477	1.478	1.481
	SO <sub>3</sub> 56.36%	1.14	0.177				
	<i>K<sub>T</sub></i>	1.201	0.1788				
Lithium sulfate (11)	Li <sub>2</sub> SO <sub>4</sub>						
	Li <sub>2</sub> O 27.18%	2.20	0.310	2.23	1.465	1.471	1.475
	SO <sub>3</sub> 72.82%	1.14	0.177				
	<i>K</i>	1.428	0.2132				

TABLE 4—(continued)

Substance	Composition		<i>k</i> values employed		Specific gravity	Average index of refraction		
			$\alpha\beta\gamma/d$	$(n-1)/d$		Experi- mental	Calculated	
							$\alpha\beta\gamma/d$	$(n-1)/d$
Arcanite (7)	K <sub>2</sub> SO <sub>4</sub>							
	K <sub>2</sub> O	54.05%	1.33	0.189	2.70	1.495	1.497	1.495
	SO <sub>3</sub>	45.95%	1.14	0.177				
	K <sub>A</sub>		1.243	0.1835				
Sodium nitrate	NaNO <sub>3</sub>							
	Na <sub>2</sub> O	36.46%	1.28	0.181	2.27	1.499	1.499	1.496
	N <sub>2</sub> O <sub>5</sub>	63.54%	1.60	0.240				
	K		1.483	0.2185				
Cesium nitrate (11)	CsNO <sub>3</sub>							
	Cs <sub>2</sub> O	72.29%	0.834	0.124	3.69	1.553	1.570	1.576
	N <sub>2</sub> O <sub>5</sub>	27.71%	1.60	0.240				
	K		1.046	0.1561				
Rubidium nitrate (11)	RbNO <sub>3</sub>							
	Rb <sub>2</sub> O	63.38%	0.885	0.129	3.12	1.518	1.529	1.529
	N <sub>2</sub> O <sub>5</sub>	36.62%	1.60	0.240				
	K		1.147	0.1697				
Borosilicate crown glass, Wright's no. 12 (11)	SiO <sub>2</sub>	72%	1.39	0.207	2.37	1.4997	1.497	1.486
	B <sub>2</sub> O <sub>3</sub>	12%	1.70	0.220				
	Na <sub>2</sub> O	11%	1.28	0.181				
	Al <sub>2</sub> O <sub>3</sub>	5%	1.38	0.193				
	K		1.415	0.2050				
Ordinary crown glass, Wright's no. 1 (11)	SiO <sub>2</sub>	74.6%	1.39	0.207	2.50	1.5055	1.510	1.510
	As <sub>2</sub> O <sub>3</sub>	0.3%	1.57	0.214				
	Mn <sub>2</sub> O <sub>3</sub>	0.1%	1.44	0.300				
	CaO	5.0%	1.63	0.225				
	K <sub>2</sub> O	11.0%	1.33	0.189				
	Na <sub>2</sub> O	9.0%	1.28	0.181				
	K		1.386	0.2040				
Sodium potassium calcium glass (11)	SiO <sub>2</sub>	61.02%	1.39	0.207	2.626	1.5528	1.555	1.545
	Na <sub>2</sub> O	6.31%	1.28	0.181				
	K <sub>2</sub> O	9.59%	1.33	0.189				
	CaO	22.78%	1.63	0.225				
	Al <sub>2</sub> O <sub>3</sub>	0.30%	1.38	0.193				
	(plus Fe <sub>2</sub> O <sub>3</sub> )							
	K		1.432	0.2077				
Barium crown glass, Wright's no. 45 (11)	SiO <sub>2</sub>	31.0%	1.39	0.207	3.54	1.6098	1.619	1.600
	B <sub>2</sub> O <sub>3</sub>	12.0%	1.70	0.220				
	Al <sub>2</sub> O <sub>3</sub>	8.0%	1.38	0.193				
	As <sub>2</sub> O <sub>3</sub>	1.0%	1.57	0.214				
	BaO	48.0%	0.91	0.127				
	K		1.198	0.1691				
MnO · SiO <sub>2</sub> glass (11)	MnO	54.15%	1.44	0.191	3.48	1.700	1.700	1.690
	SiO <sub>2</sub>	45.85%	1.39	0.207				
	K		1.417	0.1983				

TABLE 4—(continued)

Substance	Composition		<i>k</i> values employed		Specific gravity	Average index of refraction		
			$\alpha\beta\gamma/d$	$(n-1)/d$		Experi- mental	Calculated	
							$\alpha\beta\gamma/d$	$(n-1)/d$
Flint glass, Wright's no. 53 (11)	SiO <sub>2</sub>	59.3%	1.39	0.207	2.90	1.5537	1.554	1.537
	PbO	27.5%	1.08	0.137				
	K <sub>2</sub> O	8.0%	1.33	0.189				
	Na <sub>2</sub> O	5.0%	1.28	0.181				
	As <sub>2</sub> O <sub>3</sub>	0.2%	1.57	0.214				
	<i>K</i>		1.295	0.1850				
Beryllium glass, Lai and Silver- man's no. B <sub>1</sub> (11)	SiO <sub>2</sub>	73.40%	1.39	0.207	2.45	1.5193	1.506	1.501
	Na <sub>2</sub> O	18.96%	1.28	0.181				
	BeO	7.64%	1.71	0.238				
	<i>K</i>		1.393	0.2044				
Gahnite Analysis no. 11 (6)	FeO	1.71%	1.42	0.187	4.57	1.818	1.819	1.820
	MnO	0.50%	1.44	0.191				
	ZnO	41.31%	1.16	0.153				
	Al <sub>2</sub> O <sub>3</sub>	53.28%	1.38	0.193				
	Fe <sub>2</sub> O <sub>3</sub>	2.51%	2.49	0.308				
	SiO <sub>2</sub>	0.69%	1.39	0.207				
	<i>K<sub>G</sub></i>		1.318	0.1793				
Ferroan gahnite analysis no. 14 (6)	MgO	2.38%	1.46	0.200	4.38	1.782	1.793	1.789
	FeO	8.52%	1.42	0.187				
	MnO	0.10%	1.44	0.191				
	ZnO	31.32%	1.16	0.153				
	Al <sub>2</sub> O <sub>3</sub>	57.59%	1.38	0.193				
	Cr <sub>2</sub> O <sub>3</sub>	0.09%	2.09	0.270				
	<i>K<sub>G</sub></i>		1.317	0.1802				
Augite analysis no. 13 (4)	SiO <sub>2</sub>	50.91%	1.39	0.207	3.394	1.703	1.714	1.713
	Al <sub>2</sub> O <sub>3</sub>	2.68%	1.38	0.193				
	Fe <sub>2</sub> O <sub>3</sub>	1.86%	2.49	0.308				
	FeO	10.05%	1.42	0.187				
	MgO	12.39%	1.46	0.200				
	CaO	20.55%	1.63	0.225				
	Na <sub>2</sub> O	0.47%	1.28	0.181				
	K <sub>2</sub> O	0.02%	1.33	1.189				
	H <sub>2</sub> O(+)	0.19%	2.25	0.340				
	TiO <sub>2</sub>	0.40%	4.18	0.397				
	MnO	0.48%	1.44	0.191				
	<i>K<sub>A</sub></i>		1.483	0.2100				
	Hedenbergite analysis no. 18 (4)	SiO <sub>2</sub>	48.42%	1.39				
Al <sub>2</sub> O <sub>3</sub>		0.30%	1.38	0.193				
Fe <sub>2</sub> O <sub>3</sub>		1.50%	2.49	0.308				
FeO		22.97%	1.42	0.187				
MgO		1.06%	1.46	0.200				
CaO		21.33%	1.63	0.225				
Na <sub>2</sub> O		0.14%	1.28	0.181				
K <sub>2</sub> O		0.03%	1.33	0.189				
H <sub>2</sub> O(+)		0.46%	2.25	0.340				
TiO <sub>2</sub>		0.08%	4.18	0.397				
MnO		3.71%	1.44	0.191				
<i>K<sub>H</sub></i>			1.473	0.2078				

The constants are calculated (1) from the oxides themselves or (2) from compounds containing the oxides.

Indices of refraction are calculated for 26 compositions (minerals, artificial compounds, and glasses) by means of (1)  $\alpha\beta\gamma/d = k$  and (2)  $(n - 1)/d = k$ . In the majority of these examples, the former expression yields indices closer to those experimentally obtained.

In some respects the new equation is more convenient than the others. It is not necessary to compute an average index of refraction when an anisotropic substance is involved. Furthermore, if two indices are known, the third can be calculated readily, provided that  $k$  and  $d$  are known.

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# A REGULARLY INTERSTRATIFIED MONTMORILLONITE-CHLORITE

J. W. EARLEY,\* G. W. BRINDLEY,† W. J. McVEAGH,\*  
AND R. C. VANDEN HEUVEL†

## ABSTRACT

A mixed layer clay mineral having a regular alternation of montmorillonite- and chlorite-like layers is described using *x*-ray, thermal, dehydration, and chemical analysis. *X*-ray examination of the natural material shows a long spacing of about 29 Å and a fairly well developed sequence of basal spacings. The montmorillonite-like layers expand with glycerol and contract with heat treatment similar to montmorillonite while the chlorite-like layers show modified reflection intensities on heating similar to chlorite. The electron distribution normal to (001) indicates the presence of regular interstratification. Dehydration and thermal analyses yield curves which can be accounted for by attributing water loss reactions to montmorillonitic and chloritic material. The high MgO content of the clay lends support to the presence of chloritic layers. The question of nomenclature of such materials is raised.

## INTRODUCTION

The dominant clay mineral occurring in the Yates Formation, Ward County, Texas, is a mixed-layer variety having a highly regular alternation of montmorillonite- and chlorite-like layers. Core samples containing this mineral were made available for study by the Fort Worth Production Division of the Gulf Oil Corporation.

The occurrence of *randomly* stratified layer silicates is a general phenomenon found as a transition stage in the alteration of non-expanding layer silicate minerals such as micas and chlorites, to the expanding minerals montmorillonite and vermiculite, but only a few instances are known of regular interstratifications. Examples have been described by Bradley (1950)(1), Sudo et al. (1954)(2), Heystek (1954)(3), and Lippmann (1954)(4). A preliminary *x*-ray examination of the clay mineral in the Yates Formation indicated that this material might be suitable for detailed study.

## OCCURRENCE

Geologically, the clay occurs somewhat irregularly distributed in argillaceous siltstone from the Upper and Lower Yates Formation of Permian age in West Texas. The siltstone is poorly cemented and varies in color from light gray to gray-brown with numerous blue-gray streaks. Thin sections of the cores revealed quartz, clay and carbonaceous matter, feldspar, dolomite, and mica in order of abundance. The clay minerals

\* Gulf Research & Development Company, Pittsburgh, Pennsylvania.

† Pennsylvania State University, University Park, Pennsylvania.



occur dispersed throughout the rock and as cementing material, although the montmorillonite-chlorite is more abundant in the dark areas. The small platy black particles of carbonaceous matter are concentrated in, although not limited to, the blue gray streaks. Clay fractions of the silt-stone containing primarily montmorillonite-chlorite were used for the study.

### X-RAY ANALYSIS

X-ray diffractometer records of the  $<1\mu$  clay fraction in the natural and glycerol-solvated states show well-defined reflections from spacings

TABLE 1. MONTMORILLONITE-CHLORITE: X-RAY POWDER DATA  
 $<0.2\mu$  FRACTION, (FILTERED  $\text{CuK}\alpha$  RADIATION,  $\lambda$  1.5418 Å)

<i>I</i> (Est.)	<i>d</i> Å (Meas.)	<i>I</i> (Est.)	<i>d</i> Å (Meas.)
8 p	14.1	7 m	2.58
2 m, p	10.02	4 b	2.43
8 p	9.07	2 b, m	2.32
5 p	6.94		2.21
5 p	5.51	3 b, m	1.99
4 m, p	5.05	2	1.74
10	4.62	4	1.70
3 m	4.51	1 m	1.67
4	3.85	8	1.54
10 p	3.45	4 m	1.50
3 m	3.05	2	1.32
2	2.87	2 m	1.30
2	2.76		

b=broad lines.

m=mica lines.

p=preferred orientation.

of about 29 Å and 32 Å, respectively (see Fig. 1), which are assumed to be first order basal reflections. Although the diffractometer is well suited for the study of basal reflections, the powder photographic method has advantages in revealing the full diffraction pattern which is given in Table 1. However, the presence of minerals such as mica in the  $<1\mu$  fraction has precluded presentation of reliably indexed powder diffraction lines. A strong reflection at 1.542 Å and a medium strong reflection at 1.505 Å are recognized as the (060) reflections from the montmorillonite-chlorite and mica, respectively. These values lie in the ranges associated with trioctahedral and dioctahedral layer silicates. Corresponding (020) reflections are observed also, and their diffuseness points to the presence of layer displacements which are common in clay mineral structures.

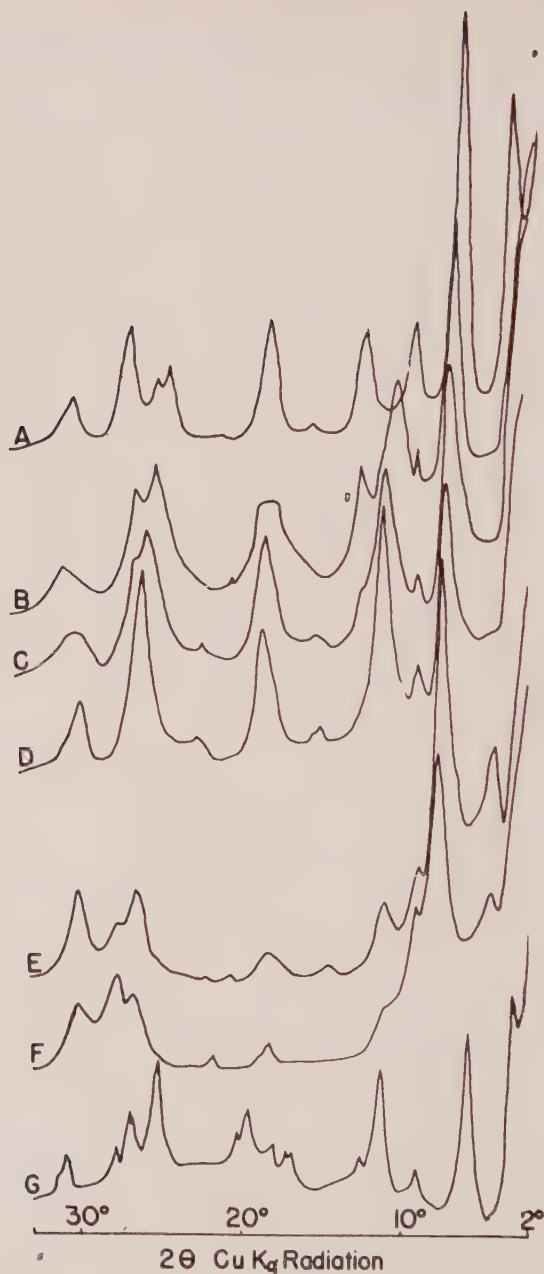


FIG. 1: X-Ray diffractometer patterns of montmorillonite-chlorite from argillaceous siltstone, Ward County, Texas, prepared from  $<.5\mu$  fraction by sedimentation on glass slides to yield maximum preferred orientation. A—Unheated, room air; B—85° C., dry air; C—200° C., dry air; D—400° C., room air; E—550° C., room air; F—700° C., room air; G—glycerol saturated, room air.

On the supposition that montmorillonite-chlorite might have hexagonal symmetry, an attempt was made to index the pattern using Bunn charts. Lack of success limited the determination of the unit cell to the  $b$  parameter:

$$b = 6 \times 1.542 = 9.25 \text{ \AA}$$

This value is comparable with the  $b$  parameters of chlorites which lie in the range 9.20 to 9.35  $\text{\AA}$ , the lower value corresponding to Mg-rich chlorite and the higher value to Fe-rich chlorite. The  $b$  parameter for Mg-montmorillonite (saponite) has been given as 9.16  $\text{\AA}$ . Thus, the  $x$ -ray data suggest that montmorillonite-chlorite is Mg-rich.

The chloritic character of the mineral was further confirmed by acid dissolution. The mineral is slightly soluble in cold 1:1 HCl but readily soluble in warm acid. Treatment with warm 1*N* HCl for a short period markedly reduced the heights of the diffractometer peaks of the montmorillonite-chlorite while the 3.53  $\text{\AA}$  peak of kaolinite and the 9.9  $\text{\AA}$  peak of mica were more clearly revealed.

Variations in the  $c$  parameter of the montmorillonite-chlorite with water loss were determined from diffractometer traces of oriented specimens in atmospheres of room air and dry air, respectively, after heating to temperatures in the range of 85° C. to 700° C. Diffractometer patterns illustrating the characteristic changes are reproduced in Fig. 1. Spacings corresponding to the reflection peaks (Table 2) are tentatively associated with integral orders of basal reflection. The mean lattice spacing derived for each temperature is no more than an approximation for a lattice rendered partially inhomogeneous by dehydration. However, the natural material in equilibrium with room air and the fully dehydrated material probably have regular layer successions.

Significant changes in the  $c$  parameter and in the reflected intensities occur on heating as shown in Table 2 and Fig. 1, respectively. In the range from 85° C. to 400° C., the basal spacings decrease from about 29  $\text{\AA}$  to 24  $\text{\AA}$  and the first order reflection disappears provided the sample is kept in dry air after heating. Under normal atmosphere conditions the mineral rehydrates very rapidly to give a first order basal spacing of about 29  $\text{\AA}$  similar to that of the unheated material. From 500 to 700° C. the first order basal reflection reappears with a spacing of about 20–22  $\text{\AA}$  while the higher orders yield a value of 23.8  $\text{\AA}$ . Thus, montmorillonite-chlorite behaves in a manner analogous to that of the chlorites in that it shows modified basal intensities after partial dehydration. In addition to these changes, the reflection peaks associated with kaolinite disappears at 600° C. confirming the presence of this mineral in the sample.

The decrease of the basal spacing from 29.1  $\text{\AA}$  to 23.8  $\text{\AA}$ , or of 5.3  $\text{\AA}$ , corresponds to a loss of roughly two water layers per unit cell. If one

chlorite layer, 14.2 Å, is subtracted from each of these parameters, the values obtained, 14.9 Å and 9.6 Å, respectively, are in good agreement with the values for air-dry and dehydrated montmorillonite. Saturated with water vapor, the mineral has a spacing of about 32 Å corresponding to approximately three layers of water.

TABLE 2. BASAL REFLECTIONS FROM NATURAL, GLYCEROL SOLVATED AND HEAT TREATED SAMPLES IN DRY AIR OF THE  $< .5\mu$  FRACTION CONTAINING MONTMORILLONITE-CHLORITE (Filtered  $\text{CuK}\alpha$  Radiation,  $\lambda 1.5418$  Å)

Natural			85° C.		200° C.		300° C.		400° C.	
<i>n</i>	<i>d/n</i>	<i>d</i> (001)	<i>d/n</i>	<i>d</i> (001)	<i>d/n</i>	<i>d</i> (001)	<i>d/n</i>	<i>d</i> (001)	<i>d/n</i>	<i>d</i> (001)
1	30.5	30.5								
2	14.5	29.0	13.5	27.0*	12.8	25.6*	12.6	25.2*	12.4	24.8*
3	9.7	29.1	8.59	25.8	8.08	24.2	8.04	24.1	7.87	23.9
4	7.25	29.0			5.87	23.5	5.91	23.6	5.91	23.6
5			4.91	24.6	4.78	23.9	4.78	23.9	4.76	23.8
6	4.85	29.1			3.95	23.7			3.88	23.3
7			3.52	24.6	3.43	24.0	3.39	23.7	3.40	23.8
8	3.64	29.1			2.94	23.5	2.92	23.4	2.96	23.7
9										
10	2.93	29.3								
Mean										
<i>d</i> (001)		29.1		25.0		23.8		23.7		23.7
500° C.			600° C.		650° C.		700° C.		Glycerol	
<i>n</i>	<i>d/n</i>	<i>d</i> (001)	<i>d/n</i>	<i>d</i> (001)	<i>d/n</i>	<i>d</i> (001)	<i>d/n</i>	<i>d</i> (001)	<i>d/n</i>	<i>d</i> (001)
1	21.6	21.6*	22.1	22.1*	21.3	21.3*	20.1	20.1*	32.7	32.7*
2	12.1	24.2*	12.0	24.0	11.8	23.6	11.6	23.2	16.1	32.2
3	7.97	23.9	7.97	23.9	8.00	24.0	8.08	24.2	9.9	29.7 m
4	5.91	23.6							8.01	32.0
5	4.77	23.8	4.87	24.3	4.84	24.2	4.86	24.3		
6									5.34	32.0
7	3.38	23.7	3.36	23.5	3.36	23.5	3.31	23.2	4.60	32.2
8	2.96	23.7	2.96	23.7	2.96	23.7	2.94	23.5		
9									3.56	32.0
11									2.91	32.0
16									1.997	31.0
Mean										
<i>d</i> (001)		23.7		23.9		23.8		23.7		32.0

\* = reflections excluded in taking mean *d*(001).

m = probably a mica reflection.

Further indication of the presence of montmorillonite layers was obtained by glycerol solvation which expands the *c*-parameter to 32.0 Å. On subtracting 14.2 Å for the chlorite, one obtains 17.8 Å for the glycerol-montmorillonite complex.

Detailed consideration of lattice structure is necessarily restricted to

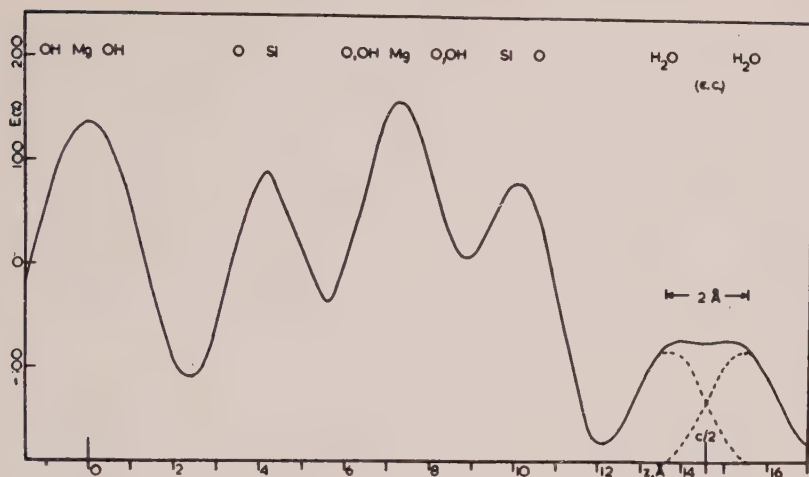


FIG. 2. Electron distribution normal to (001) of montmorillonite-chlorite.

the layer sequence in the  $c$ -direction. The calculated structure factors are given in Table 3. The reflected intensities were measured by integrating the peak areas of the diffractometer records for well-oriented layers. Structure factors were then calculated on the assumption that a well-oriented layer could be regarded as an extended face of a single crystal having mosaic texture. A counting technique to obtain more accurate

TABLE 3. COMPARISON OF  $F$  (EXP.) WITH  $F$  (CALC.) VALUES OF THE MONTMORILLONITE-CHLORITE MINERAL FROM  $<1\mu$  MATERIAL

Order of Basal Reflection	Natural			Glycerol Solvated		
	$I$	$F$ (exp.)	$F$ (calc.)	$I$	$F$ (exp.)	$F$ (calc.)
1	668	47	+35	516	49	+50
2	1,370	67	-60	616	54	-47
3	266	32	+20	16*	9	-7
4	286	39	+20	312	44	+38
5			+14			-3
6	170	42	+42	34	19	+35
7	10	10	+14	120	38	+37
8	284	56	+55			0
9	18	15	+22	298	69	+72
10	104	38	+65			+6
11				72	38	-47
16				54	42	+30

\* Mica contribution not subtracted.



intensities was not worthwhile since the reliability of the experimental  $F$  values depends on the extent of justification in applying to the oriented layer a formula derived for a mosaic crystal.

The observed and calculated  $F$  values are compared in Table 3. The observed values, which are relative, have been scaled so that  $\Sigma |F|$  is the same for the observed as for the calculated values. The agreement of the observed and calculated values leaves little doubt that the structural model is essentially correct.

Fourier syntheses of electron density were carried out using the observed  $F$  values and the calculated signs given in Table 3. The electron density curve (Fig. 2) shows clearly the alternating layers including the brucite layer of chlorite and the interlayer of montmorillonite with two layers of water. The individual atomic layers are not resolved by the Fourier synthesis because the highest order which can be recorded is 00.10 with  $d/n = 2.93 \text{ \AA}$ . The unresolved peak for the two water layers can be tentatively separated into two peaks about  $2 \text{ \AA}$  apart (see Fig. 2) and this agrees with the results of Mathieson and Walker (1954)(5) for the water layers in vermiculite; they give a separation of  $2.14 \text{ \AA}$ .

#### DEHYDRATION MEASUREMENTS

A dehydration curve of the  $<0.2\mu$  fraction of montmorillonite-chlorite was obtained with the apparatus described by Earley, Milne, and McVeagh (1953)(6) using a 0.1 gram sample of air-dry material heated at  $10^\circ \text{ C. per minute}$  to  $975^\circ \text{ C}$ . A plot of per cent water versus temperature (Fig. 3) shows two distinct water loss reactions corresponding respectively to dehydration of the brucite and octahedral layers. In addition, a low temperature loss of water is observed similar to that shown by all montmorillonite type minerals. The problem of estimating the relative proportions of montmorillonite- and chlorite-type layers in montmorillonite-chlorite from the magnitudes of the water losses is rendered difficult and inexact by the same difficulty which arises in determining the amount of structural water in a montmorillonite, namely, that a sharp distinction cannot be made between the low temperature water (mainly surface adsorbed water and water of cation hydration) and the structural or hydroxyl water associated with the octahedral layers. It can be shown, however, that the water losses are broadly consistent with the proposed 1:1 ratio of montmorillonite- and chlorite-type layers. Supposing that the low temperature water is fully driven off by  $250^\circ \text{ C.}$ , then the remaining water corresponds to 9.54 per cent of the sample weight at  $250^\circ \text{ C}$ . If  $300^\circ \text{ C.}$  is taken as the limit for low temperature water, then the remaining water corresponds to 9.26 per cent of the sample weight at this temperature. In both cases, the values derived from Fig. 3 are so close to the

theoretical value (9.65 per cent) for a 1:1 mixture of magnesian-chlorite and magnesian-montmorillonite that there can be little doubt as to the general correctness of the interpretation.

If an attempt is made to estimate the separate water losses, then we encounter again the same kind of difficulty because the dehydration curve

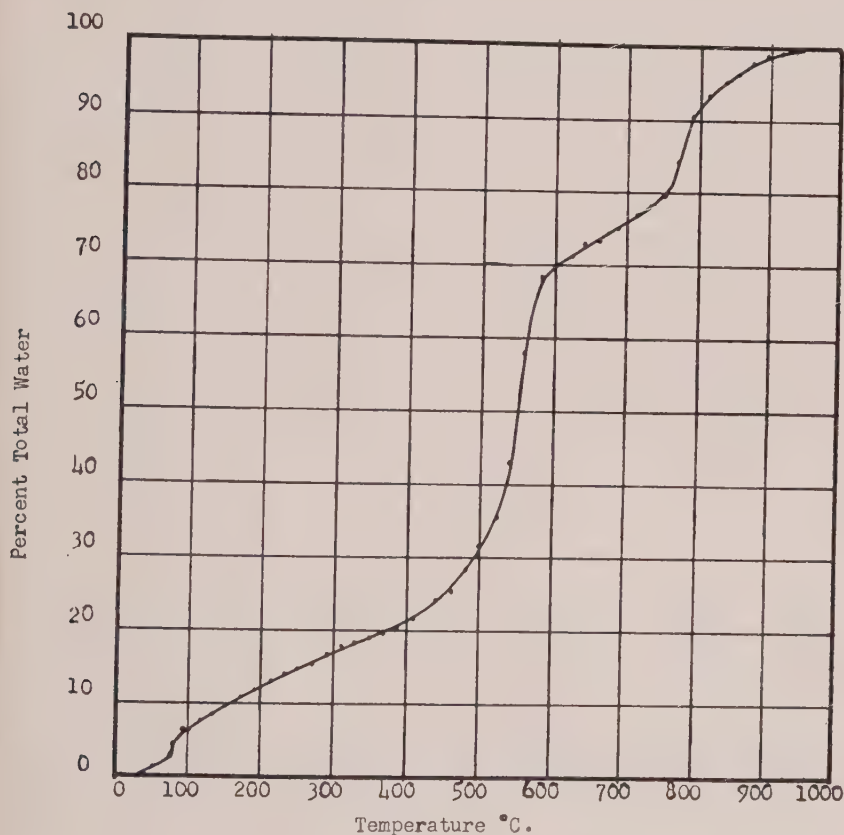


FIG. 3. Dehydration curve of montmorillonite-chlorite,  $<0.2\mu$  fraction of Yates siltstone.

does not become horizontal between the two main reactions. While the end of the first reaction can be taken fairly reliably at or near the end of the first steep rise, it is very probable that the second reaction has already commenced. The uncertainty attached to any arbitrary separation of the two water losses is such that their estimation would not provide any additional valid evidence for the 1:1 montmorillonite-chlorite sequence of layers.

## DIFFERENTIAL THERMAL ANALYSIS

Differential thermal curves were obtained by heating crude,  $<2\mu$  and  $<0.2\mu$  air-dried materials from room temperature to  $1000^{\circ}\text{C}$ . at  $10^{\circ}\text{C}$ . per minute using apparatus described by McConnell and Earley (7). Although there are minor differences in the curves from one sample to another, the dominant endothermic peaks occur at  $135^{\circ}\text{C}$ .,  $215^{\circ}\text{C}$ .,  $630^{\circ}\text{C}$ .,  $830^{\circ}\text{C}$ . and an exothermic peak at  $850^{\circ}\text{C}$ . In addition to the endotherms at  $630^{\circ}\text{C}$ . and  $830^{\circ}\text{C}$ . which are characteristic of brucite and octahedral layer water in chlorite, the reactions at  $135^{\circ}\text{C}$ . and  $215^{\circ}\text{C}$ . indicate interlayer water and water of cation hydration, respectively, similar to montmorillonite. Since the high temperature thermal effects of dioctahedral montmorillonites are missing, the montmorillonite

TABLE 4. CHEMICAL ANALYSIS\* OF THE  $<0.5\mu$  FRACTION CONTAINING PRIMARILY MONTMORILLONITE-CHLORITE

Per Cent		Per Cent	
$\text{SiO}_2$	43.1	$\text{MgO}$	17.65
$\text{Al}_2\text{O}_3$	16.6	$\text{Na}_2\text{O}$	0.52
$\text{Fe}_2\text{O}_3$	6.32	$\text{K}_2\text{O}$	2.72
$\text{TiO}_2$	0.73	$\text{CO}_2$	0.81
$\text{MnO}$	0.03	$\text{H}_2\text{O}$	7.40
$\text{P}_2\text{O}_5$	0.26	C	3.40
$\text{CaO}$	0.94	S	0.00
		Total	100.48

\* Analyst, B. B. Osthaus, Gulf Research & Development Company, Pittsburgh, Pennsylvania.

layers of montmorillonite-chlorite are probably trioctahedral as suggested by the  $830^{\circ}\text{C}$ . endotherm. The exotherm at  $850^{\circ}\text{C}$ . is attributed to the reorganization of lattice layers.

Thus, the differential thermal data substantiate the interpretation of this mineral as containing a mixture of chlorite- and montmorillonite-type layers, in agreement with the x-ray and dehydration results.

## CHEMICAL ANALYSIS

A chemical analysis of the  $<0.5\mu$  fraction of the Yates siltstone is given in Table 4.

Although some of the iron may be divalent, the total iron is expressed as trivalent because there is no analytical method for distinguishing between divalent and trivalent iron in the presence of organic matter. The  $\text{CO}_2$  content of the sample is undoubtedly due to admixed dolomite

since  $x$ -ray diffraction lines of dolomite are present. Based on this interpretation, the sample contains 1.7 per cent dolomite. The high MgO content lends support to the presence of a chlorite-type layer while the presence of 2.72 per cent  $K_2O$  suggests potash montmorillonite and/or micaceous material. Since  $x$ -ray diffraction lines of mica are present in the pattern of this fraction, the potash is very likely associated with the mica. In view of the impurities suggested by the chemical analysis and indicated by the  $x$ -ray and thermal data, calculation of a precise chemical formula for montmorillonite-chlorite would be meaningless.

### NOMENCLATURE

The question whether this mineral should have a distinctive name is worthy of consideration. Lippmann (4) has proposed the name "corrensite" for the swelling chlorite mineral which he described. However, Dr. R. C. Mackenzie, in a private communication (to G. W. B.) has expressed certain views at present under consideration by the British Clay Minerals Group† and it would be in accordance with these views to use a term such as montmorillonite-chlorite for a roughly 1:1 *random* interstratification of the two kinds of layers and a special name for a mineral in which there is a regular interstratification.

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*Manuscript received April 19, 1955.*

† These discussions have now been published. See G. Brown, Report of the Clay Minerals Group Sub-Committee on nomenclature of clay minerals, *Clay Min. Bull.*, **2**, 294-302 (1955).

## OBSERVATIONS ON HYDROHETAEROLITE

JOHN McANDREW, *Commonwealth Scientific & Industrial Research Organization, University of Melbourne, Melbourne N.3, Australia.*

### ABSTRACT

Hydrohetaerolite from Leadville, Colorado, is of tetragonal symmetry, space group  $I4/amd$ . The unit cell of dimensions  $a=5.73_5$ ,  $c=9.00_5$  Å contains  $4H\text{ZnMn}_{2-x}\text{O}_4$ , where  $x$  is approximately 0.4 and represents vacant cation positions. Decomposition into oxides of the constituent elements occurs on heating, and the recombination of these oxides to form hetaerolite,  $\text{ZnMn}_2\text{O}_4$ , at higher temperatures indicates hydrohetaerolite is a separate mineral species distinct from the former. A method of density determination for fibrous minerals is described.

A distinction between hetaerolite,  $\text{ZnMn}_2\text{O}_4$ , and the hydrous equivalent hydrohetaerolite was first made by Palache (1929), and was retained in the monograph on Franklin and Sterling Hill (Palache, 1935). An  $x$ -ray study of the two minerals has since been made by Frondel and Heinrich (1942), to which reference may be made for a revision of the relevant literature. From the hydrohetaerolite described by Ford and Bradley (1913) they obtained an  $x$ -ray powder pattern similar to that of hetaerolite, of which all but five weak lines indexed on a tetragonal cell of dimensions  $a=5.72$ ,  $c=9.05$  Å. Their  $x$ -ray single crystal photographs, stated to be of poor quality, were in agreement with the geometrically tetragonal cell. However these photographs were not sufficient to define a tetragonal symmetry, which was assumed in deducing a "partial space group"  $I4/a-d$ . Frondel (1953) has described hydrohausmannite, of composition  $\text{Mn}_3(\text{O},\text{OH})_4$ , and indicated the analogy to hydrohetaerolite.

The present investigation of hydrohetaerolite was made on a portion of the type material of Ford and Bradley (HMM92151) obtained through the courtesy of Professor Clifford Frondel. The specimen consists of a radiating aggregate of finely acicular crystals growing out from a core of finely granular crystals. The fibres show a number of cleavages parallel to the fibre axis, and are slightly twisted along this axis.

### X-RAY CRYSTALLOGRAPHY

A number of minute fibres which showed substantially no twisting were selected for the  $x$ -ray single crystal investigation. After several trials a crystal was obtained which gave small sharp spots on an  $x$ -ray rotation photograph. It was not possible to orientate the crystals by an optical goniometer to within less than  $2^\circ$  to  $3^\circ$ . Final exact orientation was therefore made on the  $x$ -ray goniometer, for which a method, essentially a variation of that described by Bairsto (1948), was developed.



Clear, sharp rotation and zero and first layer Weissenberg photographs were obtained for rotation about the axis of elongation of the fibre. The rotation photograph gave a period of  $8.12 \pm 0.01$  Å along the fibre axis, and from the zero layer Weissenberg the parameters of the rectangular lattice perpendicular to the fibre axis were measured as  $9.00_5 \pm 0.01$ ,  $8.10 \pm 0.01$  Å. The symmetry  $C_{2l}$  of both the zero and first layer Weissenberg photographs allows of tetragonal symmetry for the fibre axis the [100] or [110] directions. A Laue photograph of the fibre taken with the incident x-ray beam parallel to the direction of period  $9.00_5$  shows this

TABLE 1. X-RAY POWDER PATTERN OF HYDROHETAEROLITE,  $\text{HZnMn}_{2-x}\text{O}_4$   
Tetragonal,  $I4/amd$ ;  $a = 5.73_5$ ,  $c = 9.00_5$  Å;  $Z = 4$ . Fe radiation, Mn filter

<i>I</i>	<i>d</i> (meas.)	<i>hkl</i>	<i>d</i> (calc.)	<i>I</i>	<i>d</i> (meas.)	<i>hkl</i>	<i>d</i> (calc.)
$\frac{1}{2}$	4.82	(011)	4.837	tr	1.387	"(411)"	1.375
$\frac{1}{2}$	3.34	—	—*	$\frac{1}{4}$	1.312	(305)	1.311
7	3.02	(112)	3.013	tr	1.299	(332)	1.295
4	2.87	(200)	2.867	tr	1.283	(420)	1.282
8	2.66	(103)	2.659	3	1.263	(413)	1.262
10	2.47	(211)	2.467	$\frac{1}{2}$	1.155	(316)	1.156
1	2.25 <sub>4</sub>	(004)	2.251	$\frac{1}{2}$	1.153	—	—†
1	2.13	—	—*	tr	1.131	{(431)(501)	1.138
2	2.03	(220)	2.028			{(008)	1.126
1	1.773	(204)	1.771	$\frac{1}{2}$	1.117	(424)	1.114
2	1.723	(105)	1.718	1	1.102	(415)	1.101
2	1.686	(312)	1.682	$\frac{1}{2}$	1.093	(512)	1.092
1	1.613	(303)	1.612	3	1.074	(433)(503)	1.072
5	1.570	(231)	1.566	1	1.060	(521)	1.058
7	1.508	(224)	1.507	3	1.015	(440)	1.014
$\frac{4}{4}$	1.434	(400)	1.434	1	1.003	(523)	1.004
$\frac{1}{4}$	1.412	(314)	1.412				

\* Extraneous lines, varying in intensity among the x-ray powder patterns.

† Indexed as (334) by Frondel and Heinrich, a diffraction extinguished by the *d* glide-plane.

direction to be a fourfold axis of symmetry, and thus the tetragonal *c* axis, and establishes tetragonal symmetry. A photograph of the *hkl*0 reciprocal lattice plane obtained on a Buerger precession camera likewise shows the fourfold symmetry.

The smallest tetragonal cell has  $a = 5.73_5 \pm 0.01$  Å,  $c = 9.00_5 \pm 0.01$  Å, and for this orientation the fibre axis is the [110] direction. A partial diffraction symbol of  $I4/?md$  is given by the *hkl* diffractions present only for  $h+k=2n$  and *hhl* diffractions present only for  $2h+l=4n$  on the Weissenberg photographs. The further extinction condition of diffractions

present only for  $h=2n$  and  $k=2n$  on the  $hk0$  precession photograph leads to the complete diffraction symbol of  $4/mmmI-a-d$ , allowing  $I4/amd$  as the only possible space group. The x-ray powder pattern, Table 1, indexes for the above cell dimensions with excellent agreement between measured and calculated lattice spacings.

#### DENSITY

A determination of the density of a 5 mg. portion of the specimen was made with a Berman torsion balance. The value obtained, 4.64 gm./cc., is in close agreement with the maximum value of 4.65 measured by Frondel and Heinrich. However, it has been thought that the measurement of volume and hence of density of this fibrous material by displacement of a liquid such as toluene may be in error.

As an aid in checking the density a direct volume measurement was made of a fibre of known mass. For this determination a fibre of dimensions  $1.4 \times 0.4 \times 0.2$  mm. was selected, and its mass determined on a Becker microbalance as  $0.294 \pm 0.003$  mg. The volume of the fibre was then determined by measuring the area of parallel serial cross-sections at known distances apart. To obtain such sections the fibre was mounted in a transparent catalyst setting plastic. A reference surface was ground and polished perpendicular to the axis of the fibre. Serial sections from tip to tip of the fibre at intervals of 0.05 to 0.18 mm. were then obtained by repeated grinding and polishing on surfaces kept parallel to the reference surface. The area of each cross-section of the fibre was calculated from the area of the magnified image on the focussing screen of a Leitz Panphot microscope, the area being measured by a polar planimeter. The position of each cross-section was determined with a screw micrometer by its distance from the reference surface.

The calculation of the volume from these measurements could not be made by Simpson's rule as the sections were not spaced at equal intervals. Each cross-sectional area was therefore assumed to be the average for a segment of the fibre extending halfway to each adjacent cross-section. Calculated in this manner, the sum of the volumes of the twelve segments along the fibre is  $6.94 \times 10^{-5}$  cc., with an error estimated as not greater than 7%. This, together with the measured mass of 0.294 mg., gives a density of  $4.3 \pm 0.3$  gm./cc. Within the limits of error this determination agrees with the value of 4.64 measured on the torsion balance; the density certainly cannot be much higher, and is well below the density 5.18 gm./cc. of hetaerolite.

The errors in the direct measurement of volume arose mainly from the cross-sections being obtained by hand grinding and polishing. Although the average spacing of the cross-sections was 0.12 mm., occasional over-

grinding produced 50% larger intervals, with a marked reduction in overall accuracy of determination of the volume. Furthermore the hand polishing, completed on billiard cloth, produced distinct relief between the hydrohetaerolite and its enclosing plastic, which made the exact outline of the mineral difficult to locate. Mechanical equipment to produce constantly spaced, smaller intervals, and attainment of a relief free polish would substantially improve the accuracy of this method of density determination.

#### POLISHED SECTION

In reflected light hydrohetaerolite is creamish-gray in color, with a reflectivity estimated as 15–20% by visual comparison with minerals of known reflectivity, and is distinctly anisotropic. The absorption index of the mineral, translucent in thin flakes, is very small, and, considering it as zero, the reflectivities in polished section calculate as  $R_0 = 15.0\%$ ,  $R_e = 12.5\%$ , from the refractive index measurements  $n_o = 2.26$ ,  $n_e = 2.10$  of Berman (Palache, 1929). In comparison the reflectivities of hetaerolite calculate as  $R_0 = 16.1\%$ ,  $R_e = 13.1\%$  in good agreement with the values 17.7%, 13% measured by Orcel and Pavlovitch (1931) in white light. The properties in reflected light correspond closely with the description of hetaerolite from Sterling Hill by Orcel and Pavlovitch. However hydrohetaerolite is tarnished by 30%  $\text{HNO}_3$ , which does not affect hetaerolite although conc.  $\text{HNO}_3$  has a positive reaction with the latter.

#### HEAT TREATMENT

There is a clear although slight distinction in physical properties between hydrohetaerolite and hetaerolite. The essential difference in chemical composition is the presence of about 4% water in the former. A series of heating experiments was therefore made to find if hydrohetaerolite transformed to hetaerolite by loss of water at elevated temperatures.

Powdered 0.2 mg. portions of the Leadville hydrohetaerolite were heated in air for 2 hours at  $400 \pm 5^\circ \text{C}$ .,  $600 \pm 5^\circ \text{C}$ ., and  $860 \pm 30^\circ \text{C}$ . Heating at  $400^\circ \text{C}$ . produces no change in the x-ray powder pattern or in appearance. The product of heating at  $600^\circ$  is however distinctly lighter brown in color than the initial dark brown powder. The x-ray powder pattern reveals decomposition of the hydrohetaerolite to manganosite,  $\text{MnO}$ , and zincite,  $\text{ZnO}$ , together with the formation of a small amount of hetaerolite. Although  $\text{Mn}_2\text{O}_3$  is the stable oxide of manganese at this temperature (Mason, 1943) none of its diffraction lines are present in the x-ray pattern. A portion of this mixture was heated for 2 hours at  $850 \pm 5^\circ \text{C}$ . in air. An x-ray pattern shows that most of the  $\text{ZnO}$  and

MnO of the mixture combines at the higher temperature to form hetaerolite.

The product of heating at  $860^\circ$  gave a strong x-ray powder pattern of hetaerolite, together with a few additional, weak lines. This pattern indexes for a tetragonal cell with  $a=5.73$ ,  $c=9.24$  Å, in comparison with the parameters 5.72, 9.24 measured for pure artificial  $\text{ZnMn}_2\text{O}_4$  by Mason (1947), and 5.75, 9.17 obtained for Sterling Hill hetaerolite by Frondel and Heinrich.

Thus in air hydrohetaerolite remains unaltered at temperatures of  $400^\circ$  and lower. Above a temperature between  $400^\circ$  and  $600^\circ$  it decomposes to a mixture of ZnO and MnO. At  $600^\circ$ , and more readily at higher temperatures, this mixture of oxides combines to form anhydrous  $\text{ZnMn}_2\text{O}_4$ , hetaerolite. Hydrohetaerolite thus has a different stability range from that of hetaerolite, and is a solid phase distinct from the latter. Accordingly, it is to be regarded as a separate mineral species distinct from the anhydrous hetaerolite.

#### UNIT CELL CONTENTS

The close similarity of the x-ray powder patterns and cell dimensions of hydrohetaerolite with those of hetaerolite and hausmannite, all of the same space group, shows that these minerals are isostructural. The atomic contents of the unit cell should accord with the ideal structure determined for hausmannite by Aminoff (1926), which contains 16 oxygen ions approximately in cubic close packing in this cell. There is thus not sufficient space to geometrically fit more than 16 oxygen ions into the cell of hydrohetaerolite, which is distinctly smaller than that of hausmannite. Moreover the symmetry of the space group is such that each set of equivalent positions in the unit cell contains either four atomic positions or a multiple thereof.

The cell contents calculated for the measured density are listed in Table 2 for the analysis by Bradley, 1, Bradley's analysis assuming the  $\text{SiO}_2$  present is due to admixed hemimorphite, 2, and for the analysis by Palmer 3 (Wells, 1937). In the second calculation the density is taken as 4.85 gm./cc. in allowance of the assumed 10.7% of hemimorphite of density 3.50. The oxygen content of the unit cell is close to 16, and the cell contents and required density for 16.00 oxygen ions in the cell are also given.

The unit cell contains close to  $\text{H}_4\text{Zn}_4(\text{Mn, Fe, Si})_8 \cdot \text{O}_{16}$ . The structural formula of hydrohetaerolite is thus  $\text{HZnMn}_{2-x}\text{O}_4$ , with  $x$  about 0.4, and the presence of a deficiency of cations is revealed. A similar deficiency has been found in artificial hausmannite-like preparations by Dubois (1934), who prepared a compound of composition  $\text{Mn}_2\text{O}_3$  but with an x-ray

pattern closely resembling hausmannite,  $\text{Mn}_3\text{O}_4$ . Verwey and de Boer (1936) showed this can be described as  $\text{Mn}_{3-1/3}\text{O}_4$  with 4/3 vacant cation positions per unit cell, analogous to  $\gamma\text{-Fe}_2\text{O}_3$ . The analysis of hydrohetaerolite indicates a greater proportion of vacant cation positions.

A variably hydrated manganese oxide isostructural with hausmannite has been prepared by Dubois (1934) and by Feitknecht and Marti (1945), and occurs naturally as the mineral hydrohausmannite, Frondel

TABLE 2. ANALYSES AND UNIT CELL CONTENTS OF HYDROHETAEROLITE  
ANALYSES

	1	2	3
ZnO	37.56	30.30	37.66
Mn <sub>2</sub> O <sub>3</sub>	—	—	54.93
MnO	50.34	50.34	—
O	5.99	5.99	—
Fe <sub>2</sub> O <sub>3</sub>	—	—	0.67
SiO <sub>2</sub>	2.69	—	2.91
H <sub>2</sub> O	4.36	3.55	3.78
Hemimorphite	—	10.76	—
Total	100.94	100.94	99.65
G(meas.)	4.64	4.85	4.64

CALCULATED UNIT CELL CONTENTS			UNIT CELL CONTENTS FOR O=16			
	1	2	3	1	2	3
Zn	3.80	3.58	3.86	3.91	3.47	4.07
Mn	5.84	7.42	5.77	6.00	7.19	6.08
Fe	—	—	0.07	—	—	0.07
Si	0.39	—	0.40	0.40	—	0.41
H	3.98	3.80	3.50	4.10	3.68	3.68
O	15.55	16.50	15.17	16.00	16.00	16.00
G(calc.)				4.77	4.72	4.89

1. Hydrohetaerolite, Leadville, Colorado; Ford and Bradley (1913).
2. The same, considering the  $\text{SiO}_2$  is in admixed hemimorphite.
3. Hydrohetaerolite, Leadville, Colorado; Palmer (Wells, 1937).

(1953). In analogy with the structure of hausmannite, the unit cell of hydrohausmannite contains  $16(\text{O},\text{OH})$ . A higher anion content is geometrically improbable. Moreover Verwey and de Boer (1936) show that the structure and electrical conductivity of hausmannite indicate  $\text{Mn}^{2+}\text{Mn}^{4+}$  as the cations rather than  $\text{Mn}^{2+}\text{Mn}^{3+}_2$ . On this basis the analysis of hydrohausmannite (backstromite) from Långban, Sweden, by Mauzelius (Aminoff, 1919) gives a unit cell contents of  $4 \times (\text{Mn}^{2+}$ ,



$\text{Mg}_{1.42}\text{Mn}_{4.00}\text{O}_{4.00}\text{H}_{1.16}$ , excluding  $\text{H}_2\text{O}-130^\circ$ . The alternative cation valencies correspond to a cell contents of  $4 \times (\text{Mn}^{2+}, \text{Mg})_{0.42}\text{Mn}_{3.20}\text{O}_{4.00}\text{H}_{1.16}$ . The density of hydrohausmannite with these atomic contents in a cell of dimensions  $a=5.79$ ,  $c=9.49$  Å (Fron del, 1953), calculates as 4.13 gm./cc., much below the density 4.84 gm./cc. of hausmannite.

The difference in cell dimensions of hydrohetaerolite and hetaerolite is greater than the differences in the variants of hausmannite, Table 3.

TABLE 3. CELL DIMENSIONS OF HYDROHETAEROLITE AND RELATED STRUCTURES

	<i>a</i>	<i>c</i>	Cell Contents/4	Reference
Hydrohetaerolite	5.73 <sub>5</sub>	9.00 <sub>5</sub>	$\text{HZnMn}_{2-0.4}\text{O}_4$	(Present study)
Hetaerolite	5.73	9.24	$\text{ZnMn}_2\text{O}_4$	(Present study)
Hydrohausmannite	5.79	9.49	$\text{Mn}_{3-0.6}(\text{O}, \text{OH})_4$	Fron del (1953)
Hausmannite	5.79	9.44	$\text{Mn}_3\text{O}_4$	Aminoff (1926)
$\gamma\text{-Mn}_2\text{O}_3$	5.7	9.4	$\text{Mn}_{3-1/3}\text{O}_4$	Verwey and deBoer (1936)
Magnetite	5.94 <sup>1</sup>	8.40	$\text{Fe}_3\text{O}_4$	Hagg (1935)
Maghemite	5.90 <sup>1</sup>	8.34	$\text{Fe}_{3-1/3}\text{O}_4$	Hagg (1935)

<sup>1</sup> Dimensions of the structural unit which corresponds to the unit cells of the other minerals, and is half the volume of the cubic cell.

The slight increase in cell dimensions of hydrohausmannite relative to hausmannite, and the strong (002) diffraction in the x-ray powder pattern of the former have no parallel in hydrohetaerolite. The presence of cation vacancies in hausmannite and in magnetite, of similar structure to hetaerolite, results in only a slight decrease in the cell dimensions.

In structures of the hausmannite type (Aminoff, 1926) the oxygen ions are in a distorted cubic close-packing. The oxygen ions in layers parallel to 001 are in a close packed arrangement with but little separation beyond the normal ionic radius, and the distortion is in the *c* axial direction as a separation of these layers beyond the ionic distance in the 001 planes. This distortion is distinctly smaller in hydrohetaerolite than in hetaerolite, and the reduction may arise from the position of the hydrogen in the structure, and the nature of the hydrogen bond.

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## CRYSTALLOGRAPHY AND COMPOSITION OF CREDNERITE

JOHN McANDREW, *Commonwealth Scientific and Industrial Research  
Organization, University of Melbourne, Melbourne N.3, Australia.*

### ABSTRACT

Crednerite from the Mendip Hills, Somerset, is monoclinic, with unit cell dimensions  $a$  5.58,  $b$  2.87,  $c$  5.87 Å,  $\beta$  104.0°, and space group  $C2/m$ ,  $C2$  or  $Cm$ . The composition of  $\text{CuMn}_2\text{O}_4$ , from the chemical analysis by Mountain, gives an irrational unit cell content for the measured density of 5.03 gm./cc. Above 1100° C. in air copper manganese oxide mixtures form crednerite together with copper hausmannite or cuprite, depending on the overall composition. The synthetic crednerite is of composition  $\text{Cu}_2\text{Mn}_2\text{O}_5$ , comparable to crednerite from Friedrichroda.

Crednerite was first found as foliated crystals at Friedrichroda, Thuringia, from where it was described by Credner (1847). A perfect basal cleavage with two less perfect cleavages oblique to it indicated a possible monoclinic symmetry, and Rammelsberg (1849) deduced the chemical formula of  $3\text{CuO} \cdot 2\text{Mn}_2\text{O}_3$  from analyses and gave to this mineral the name crednerite. Meanwhile Hausmann (1847) had briefly described it in Vol. 2 of his "Handbuch der Mineralogie" (p. 1582) under the name mangankupferoxyd.

No other occurrence of crednerite was described until 1923, when Spencer and Mountain (1923) identified it as plates up to  $10 \times 10 \times 1$  mm. from a locality at the Higher Pitts Farm, Priddy, Mendip Hills, Somerset. The broad surface of the plates showed three sets of striae and overlapping portions at angles of 56° to 63°, suggesting twinning. Cleavage at the edges was parallel to the striae and inclined at 76° to the cleavage parallel to the plates, again suggesting a possible monoclinic symmetry with the pseudo-hexagonal appearance arising from twinning. The chemical analysis by Mountain was in very close agreement with the formula  $\text{CuMn}_2\text{O}_4$ .

### DESCRIPTION

For the present investigation some plates of crednerite from the Mendip Hills were made available by the courtesy of the British Museum (Natural History) (BM 1923, 729). These correspond in physical properties, including form and cleavage, with the description of Spencer and Mountain. Two specimens from Friedrichroda labelled crednerite were examined but each was discovered to be incorrectly identified. The crystallographic study was therefore confined to the material from the Mendip Hills.

This crednerite consists of bright gray plates with a metallic lustre

which are striated on the broad surfaces in three directions. The angles between the striae in the plane of the plates were measured under a microscope as either about  $55^\circ$  or  $69^\circ$ . Cleaving along the perfect cleavage parallel to the plates produces flat highly reflecting surfaces which are unstriated. The plates are quite brittle, readily breaking parallel to the striations to produce very thin blade-shaped cleavage fragments.

A spectrographic analysis was made of an 0.2 mg portion of the material. The powdered sample, mixed with 2 mg. of high purity carbon, was volatilized in the anode of a carbon arc on a Hilger Medium Quartz spectrograph. The spectrogram showed Cu and Mn as the two major elements, confirming the mineral as crednerite.

Under an ore-microscope the cleavage planes of crednerite are cream-white in color, with a reflectivity between 30% and 40%, estimated by visual comparison with minerals of known reflectivity. The prominent (001) cleavage is distinctly anisotropic in polarized light, and sometimes shows a mosaic intergrowth of two or three twin components each with a sharp but different extinction position. A slight tarnish was obtained with 20% HCl, while 30%  $\text{HNO}_3$ , 20% KCN, 20%  $\text{FeCl}_3$ , and saturated KOH and  $\text{HgCl}_2$  had no effect after 1 minute. These properties observed in reflected light correspond to those given by Orcel and Pavlovitch (1932) for crednerite from Friedrichroda, except that the latter shows a non-persistent tarnish with 20%  $\text{FeCl}_3$ .

#### CRYSTALLOGRAPHY

Sharp, clear rotation, and zero and first layer Weissenberg photographs were obtained with Cu radiation from a cleavage fragment of approximately square cross-section, for rotation about what was found to be the monoclinic  $b$  axis. Another set of single crystal photographs taken of a blade-shaped cleavage fragment show a pronounced diminution of the diffraction intensities in portions of the films. This is attributed to varying absorption of the  $x$ -ray beam within the fragment due to its much elongated cross-section, in the manner described by Wells (1937).

From the rotation photograph a lattice period of  $2.87_7 \pm 0.005 \text{ \AA}$  was calculated for the axis of rotation. Both zero and first layer Weissenberg photographs show a plane point group symmetry of  $C_2$ , for which the axis of rotation is either the [010] direction in a crystal of monoclinic symmetry, or the  $[10\bar{1}0]$  direction in a crystal of rhombohedral diffraction symmetry  $\bar{3}m$ . The latter requires a rectangular cell in the zero and first layer, and in fact a small rectangular cell can be chosen on the reciprocal lattice sections plotted from the Weissenberg photographs. One period in this rectangular reciprocal plane lattice should then cor-

respond to the spacing of the  $(10\bar{1}0)$  planes in the hexagonal lattice, perpendicular to the axis of rotation. The reciprocal lattice spacing of these planes was determined from the rotation photograph as 0.62 on a scale of  $\lambda/d$ ; the rectangular reciprocal lattice was however of dimensions  $0.27 \times 0.07$ , so the rhombohedral possibility was discarded.

The smallest monoclinic cell with least obliquity was chosen, and its dimensions determined as

$$\begin{aligned} a &= 5.58 \pm 0.01 \text{ \AA} \\ b &= 2.87_7 \pm 0.005 \text{ \AA} \\ c &= 5.87_6 \pm 0.01 \text{ \AA} \\ \beta &= 104.0^\circ \pm 0.1^\circ \end{aligned}$$

The only systematic extinction conditions for this cell are in  $hkl$  diffractions, present only when  $h+k$  is even. The cell is thus  $C$  centered, and the possible space groups are  $C2/m$ ,  $C2$  or  $Cm$ .

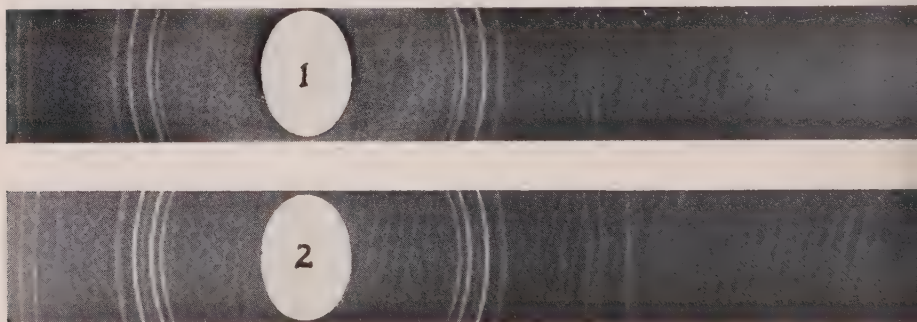


FIG. 1. X-ray powder patterns of 1: crednerite, Mendip, Hills  
2: artificial  $\text{Cu}_2\text{Mn}_2\text{O}_6$ .

The x-ray powder pattern, Table 1, was indexed for this cell, and the agreement between observed and calculated spacings confirms both the diffraction lines of this pattern and the cell dimensions calculated from the single crystal photographs.

The lattice of crednerite also contains a monoclinic cell which has  $\beta = 93^\circ$ , and is thus geometrically pseudo-orthorhombic. This cell is of dimensions  $a' = 7.05_6 \text{ \AA}$ ,  $b' = 2.87_7 \text{ \AA}$ ,  $c' = 9.03 \text{ \AA}$ ,  $\beta' = 93.0^\circ$ , and is twice the volume of the smaller  $C$ -centered cell. The orientation of the pseudo-orthorhombic cell is obtained from that of the unit cell selected by the transformation  $10\bar{1}/010/10\bar{1}$ .

#### CLEAVAGE

Twelve cleavage fragments were measured on a two-circle optical goniometer. In general the cleavage faces gave reflections of poor quality.



TABLE 1. CREDNERITE: X-RAY POWDER DATA  
Fe radiation, Mn filter

Cu <sub>2</sub> Mn <sub>2</sub> O <sub>8</sub> <sup>1</sup> Synthetic		Crednerite Mendip Hills			
<i>I</i>	<i>d</i> (meas.) Å	<i>I</i>	<i>d</i> (meas.) Å	<i>hkl</i>	<i>d</i> (calc.) Å
6	2.85	6	2.85	(002)	2.85
9	2.67	10	2.71	{(200)	2.71
2	2.54	2	2.54	{(201)	2.71
10	2.43	10	2.43	(110)	2.54
2	2.25			(111)	2.43
3	2.22	5	2.24 br.	{(202)	2.25
1	1.897			{(201)	2.24
2	1.745			{(111)	2.23
3	1.615	$\frac{1}{2}$	1.895	(003)	1.900
$\frac{1}{2}$	1.582	3	1.764	{(203)	1.769
3	1.552	4	1.613	{(202)	1.762
3	1.441			(113)	1.612
$\frac{1}{2}$	1.427	$\frac{1}{4}$	1.584		
$\frac{1}{2}$	1.414	4	1.560	(311)	1.559
$\frac{1}{4}$	1.395	3	1.446	(020)	1.439
$\frac{1}{4}$	1.348		—		
1	1.333	2	1.409	{(204)	1.410
1	1.287		—	{(311)	1.406
2	1.269	$\frac{1}{2}$	1.359	{(402)	1.359
1	1.216	$\frac{1}{2}$	1.330	{(400)	1.354
$\frac{1}{4}$	1.173	$\frac{1}{2}$	1.287	(313)	1.332
$\frac{1}{2}$	1.146br.	$\frac{1}{2}$	1.273	(022)	1.284
1	1.128	1	1.273	{(221)	1.271
3	1.112	$\frac{1}{4}$	1.214	{(220)	1.270
1	1.091		—	{(222)	1.213
1	1.080		—	{(221)	1.211
$\frac{1}{2}$	1.068	$\frac{1}{4}$	1.129	{(402)	1.129
2	1.033	$\frac{1}{2}$	1.117	{(404)	1.127
1	1.024	1	1.091	(223)	1.116
1	1.015			{(313)	1.087
3	1.011			{(115)	1.087
2	0.998	1	1.043	(511)	1.039
$\frac{1}{2}$	0.993	$\frac{1}{2}$	1.009	{(224)	1.007
$\frac{1}{2}$	0.987			{(223)	1.005
$\frac{1}{2}$	0.980	1	0.989	{(422'}	0.987
				{(420'}	0.986

<sup>1</sup> Plus four weak lines 4.88— $\frac{1}{2}$ , 3.02—1, 2.48—1, 2.03— $\frac{1}{2}$  from copper hausmannite.

The measurements reveal three prominent cleavage planes oblique to the cleavage in the plane of the plates. Two pairs of additional, poorer cleavage planes are also present in some fragments.

The angular positions of the cleavage planes are in accordance with the monoclinic symmetry of the single crystal  $x$ -ray photographs, but also show some pseudo-orthorhombic characteristics. From an instrumental correlation between the two-circle optical goniometer measurements and the diffractions recorded on the zero layer Weissenberg photograph, two of the prominent cleavages were identified as (001) parallel to the plane of the plates, and (100) oblique to this plane. By comparison with the crystallographic angles calculated from the unit cell dimensions, the

TABLE 2. CREDNERITE: MEASURED AND CALCULATED CLEAVAGE ANGLES\*

Cleavage	Measured		No. of faces	Calculated	
	$\phi_2$	$\rho_2$		$\phi_2$	$\rho_2$
001:perfect	76°01'† (74°30'–78°10')	88°40'–90°	12	76°00'	90°00'
100:perfect	0°00'	88°30'–91°40'	12	0°00'	90°00'
$\bar{1}11$ :good	121°–140°	32°–38°	5	129°54'	32°33'
111:poor	31°–38°	36°–42°	4	36°50'	39°14'
212:very poor	32°–37½°	45½°–47½°	2	36°50'	49°19'

\*  $\phi_2$  and  $\rho_2$  are given for the second setting of Palache, Berman, and Frondel (1944, p. 18) in the monoclinic system, in which the monoclinic  $b$  axis is vertical, with  $\rho_2$  the angle from 010 and  $\phi_2$  measured positive clockwise from 100 in the direction of 001.

† Measurement from two cleavage fragments with sharp reflections from both (001) and (100).

other two prominent cleavage planes were identified as ( $\bar{1}11$ ); and in addition (111) as a poor cleavage, and (212) a doubtful cleavage observed as two reflections of very poor quality from only one fragment. The range of angles obtained from the better reflections, together with the angles calculated from the cell dimensions, are listed in Table 2.

#### TWINNING

The intergrowth seen in reflected polarized light is undoubtedly due to twinning. On one fragment, each of the three components  $A$ ,  $B$ ,  $C$ , corresponds to one of the three directions of striae present on the 001 crystal face. On any one part of this surface striae are present in one direction only, and the border marking change of direction of the striae corresponds exactly with the boundaries of the intergrowth observed in reflected polarized light.

The nature of the twinning was determined by measuring the cleavage of this fragment with a two-circle goniometer, distinguishing the cleavage planes on each of the three components. On a stereographic projection, Fig. 2, it is seen that the adjacent components *A* and *B* are in a definite crystallographic relationship; for within the accuracy of the measurements the  $41\bar{1}$  planes of each component are parallel, and on rotation of *A* an angle of  $180^\circ$  around the normal to its  $41\bar{1}$  plane the exact orientation of the component *B* is obtained. The adjacent components *B* and *C* of the fragment are in similar twin relationship, the  $41\bar{1}$  planes of each

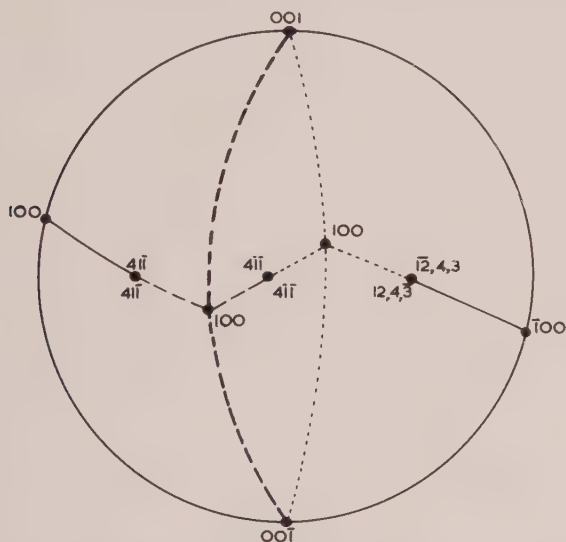


FIG. 2. Stereographic projection of twinned crystal of crednerite. (001) is common to the three components, distinguished by the solid and coarse and fine dashed lines. The indices of faces which are parallel in the two components are given on the appropriate side for each component.

being parallel, and the orientation of one related to that of the other by a  $180^\circ$  rotation around the normal to this plane. The components *C* and *A* of the twin, although actually related in orientation through the component *B*, form a pseudotwin, in which the orientation of one may be derived from that of the other by a  $180^\circ$  rotation about a line inclined at  $2\frac{1}{2}^\circ$  to the normal to the  $12, 4, 3$  plane.

The twinning conforms to the normal type of rotation twin in which the twin axis is normal to a possible face of the crystal. In some of the crednerite plates 001 appears to be a composition plane, and in one fragment the (100) cleavage is stepped parallel to the 001 plane, supporting

this observation. Nevertheless, in reflected light the twinned components are seen to be complexly intergrown on some (001) surfaces.

The prominent 001 face, parallel to the broad surface of the crednerite plates, is almost parallel in both components of a crystal twinned on this (41 $\bar{1}$ ) law. However the intersection of (100) with (001), which is the direction [010] of the striae on the broad faces of the plates, is rotated an angle of 56° in one twin component relative to the other. In a crystal twinned twice on this law, the three directions of striae on the parallel (001) faces are at angles of 56°, 56° and 68°, producing the pseudo-hexagonal appearance which was noted by Spencer and Mountain.

#### TWINNING FROM X-RAY SINGLE CRYSTAL PHOTOGRAPHS

An intergrowth with the relative orientation of the pseudotwin was also found on one set of the single crystal *x*-ray photographs. One rotation photograph about the *b* axis of a blade-shaped cleavage fragment has ten closely spaced layer lines, of which the zero and sixth are far stronger than the others, and coincide with the zero and first layers normally obtained on rotation about this axis of period 2.87 Å. In addition, the central part of some of the layer lines have no diffraction spots, and this gap varies systematically from a maximum for the third and eighth layers to nil for the zero, fifth and sixth layers. The gap in the third layer line extends for two-thirds the width of the rotation photograph, which records the complete cone of diffractions of each layer except for a small gap of 1/15 of the circumference of each cone at intersection with the film. The spacing of the layers corresponds to a real lattice period of 17.3 Å along the axis of rotation.

A Weissenberg resolution of the zero layer gave the normal *h*0*l* reciprocal plane lattice of crednerite. However, the weak first layer gave diffractions on a Weissenberg photograph corresponding to only a single line in the reciprocal lattice, with a repetition unit equal to *c*\*. This line parallel to *c*\*, which constitutes the first layer line, is a distance determined as 0.60 from the central 000 point of the reciprocal lattice at a scale of  $\lambda/d$ , and at a height 0.089 above the zero level. A cross-section of the reciprocal lattice of crednerite perpendicular to the *c*\* axis, Fig. 3, shows that the four lines 1 and 5 are the only reciprocal lattice lines parallel to *c*\* which are a distance 0.60 from the origin and contain points corresponding to possible *hkl* diffractions. A small portion of the crednerite cleavage fragment used for this set of single crystal photographs was therefore in an orientation such that one of these four lines, which are crystallographically equivalent, was parallel to the plane of the zero-level diffractions and a distance 0.089 from this plane. The zero-level must therefore be one of the planes perpendicular to the cross-section,

Fig. 3, and intersecting it along  $AA'$ ,  $BB'$ , or the equivalent directions  $A_1A_1'$ ,  $B_1B_1'$ .

Consider the rotation photograph of crednerite oriented with the axis of rotation perpendicular to  $AA'c^*$ . Reciprocal lattice points lie along lines 0, 1, 2, 3, & 3', 4,  $\dots$ , parallel to the zero layer and at equal increments of 0.089 from it. The corresponding diffractions fall along layer lines spaced according to a real lattice period of  $17.3 \text{ \AA}$ ; and each layer (except one) arises from one reciprocal lattice line. The reciprocal lattice

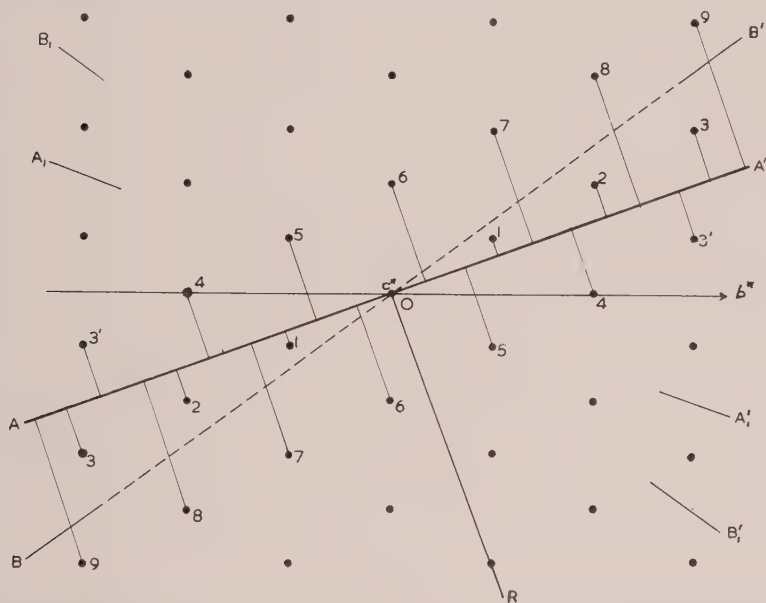


FIG. 3. Section through the centre of the reciprocal lattice of crednerite perpendicular to  $c^*$ . Dots mark the position of lines parallel to  $c^*$  in the reciprocal lattice which contain the points, with  $h+k=2n$ , for which diffractions may occur.

lines corresponding to the third and ninth layers are distant from the axis of rotation, and, with all points having high values of  $\xi^1$ , diffractions appear only at each extremity of the layer line. The other lines are at smaller distances from the axis of rotation, and have smaller central gaps in the corresponding layer line on the rotation photograph. These gaps exactly correspond to, and account for, those observed in the rotation photograph and described above.

The measurements on the rotation and Weissenberg photographs also allow the plane  $BB'c^*$  as zero-layer at a distance 0.089 from line 1 in the

<sup>1</sup>  $\xi$ : The distance, in the reciprocal lattice, from the axis of rotation.



reciprocal lattice. However, the distances from this plane to the reciprocal lattice lines parallel to  $c^*$  do not correspond to the layer-line spacings, so this orientation is rejected.

The interpretation of these single crystal photographs determines the relative orientation of the two components giving the strong and weak layers respectively. The intergrowth is such that the 001 planes of each component are parallel, and the directions [010] of the major component, and [320] of the minor component, normal to  $4.1'c^*$ , are parallel to the axis of rotation. The orientation of one component is obtained from that of the other by rotation of  $180^\circ$  about a line inclined from the normal to the  $1\bar{2}, 4, 3$  plane, towards  $b$  at  $2\frac{1}{2}^\circ$ ; the relationship found by optical goniometer measurements between two components each actually twinned with a third component by  $180^\circ$  rotation about the normal to  $41\bar{1}$ .

The interpretation of the rotation photograph also provides examples of a layer-line composed of diffractions corresponding to only a single line in the reciprocal lattice. These diffractions form prominent layer-lines when the axis of rotation is slightly inclined to a direction of small repetition distance in the crystal lattice, and is also perpendicular to a line of small repetition distance in the reciprocal lattice—thus being parallel to a crystallographic plane with low  $hkl$  indices. In the example described the axis of rotation, *OR* in Fig. 3, lies in the 001 plane and is inclined at  $8^\circ$  to [110]. Rotation about an axis in 001 at any lower inclination to [110] would likewise give layer lines, corresponding to the reciprocal lattice lines parallel to [001], although there is no lattice row in the crystal along the axis of rotation. Such rotation photographs would show, relative to that for  $8^\circ$  inclination, a splitting of the third layer line into two layer lines, and have layer lines in two groups, three more closely spaced on each side of the zero layer, and three more closely spaced on each side of the former sixth layer. The layer lines within these two groups become progressively closer with decreasing inclination, and coincide to form the zero and first layers respectively for rotation about [110], at zero inclination.

The occurrence of such layer lines, *without* a zone axis and lattice row of the crystal parallel to the axis of rotation would generally be revealed by a non-regularity in spacing of the outer layer lines relative to the inner, or by the absence of spots in the front diffraction portion of some layer lines. The spacing of such layer lines is not a measure of a lattice period along the axis of rotation. In addition, the normal explanation of layer lines as diffractions restricted to cones around the axis of rotation, the angle of the cones determined by the lattice period along this axis and the  $x$ -ray wavelength, does not apply.

## UNIT CELL CONTENTS

Crednerite from the Mendip Hills has been analyzed by Mountain (Spencer and Mountain, 1923). Although the analysis, as corrected for 1.05% cerussite and 6.10% malachite, gives a Cu:Mn:O ratio close to 1:2:4, the calculated cell contents, Table 3, are quite irrational. The

TABLE 3. CREDNERITE—ANALYSIS AND UNIT CELL CONTENTS

	1	2		3
MnO	54.40	58.62	Mn	2.29
CuO	36.57	34.68	Cu	1.16
O	6.22	6.70	O	4.66
H <sub>2</sub> O	1.88	—		
CO <sub>2</sub>				
PbO	0.88	—		
Total	99.95	100.00	D(meas.)	5.03

1. Crednerite, Mendip Hills, analysis of Mountain (Spencer and Mountain, 1923).
2. The same, after deducing 1.05% cerussite, 6.10% malachite, and recalculation to 100%.
3. Atomic contents of the unit cell, calculated for density of 5.03.

closest rational cell contents of  $\text{CuMn}_2\text{O}_4$  requires a density of 4.32 gm./cc., which is far below the value 5.03 obtained by Mountain for crednerite from the Mendip Hills, and the densities of 4.96 to 5.03 given by Credner (1847, 1848) and by Rammelsberg (1849) for crednerite from Friedrichroda.

## SYNTHESIS

The seven analyses of crednerite from Friedrichroda published by Credner (1847, 1848) and by Rammelsberg (1849) show wide variation. Omitting a small BaO content, two of the analyses correspond closely to the composition  $\text{CuO} \cdot \text{Mn}_2\text{O}_3$  of crednerite from the Mendip Hills. Analyses of higher copper content correspond to compositions close to  $3\text{CuO} \cdot 2\text{Mn}_2\text{O}_3$  and to  $4\text{CuO} \cdot 3\text{Mn}_2\text{O}_3$ , and one analysis corresponds to a composition of  $2\text{CuO} \cdot 3\text{Mn}_2\text{O}_3$ . Because of this variation of analyses, and the irrational unit cell contents obtained from the crystallographic study, synthetic crednerite was prepared and analyzed.

Intimate mixtures of copper and manganese oxides were prepared by adding a solution of the sulfates to an excess of NaOH solution, which gave complete precipitation of the copper and manganese. After washing with hot distilled water, the precipitates were air dried at 150° C., finely ground, and then shaken with four steel balls in 1 oz. bottles for five minutes to ensure complete mixing. The resulting powders were briquetted into small tablets in a hydraulic press.

Tablets with Cu:Mn=1:2 were heated on platinum foil at temperatures of 1100° to 1300° C. for 1 to 24 hours, and then quenched in air. In polished section these tablets are completely crystalline, consisting of intergrowths of copper hausmannite and crednerite. The hausmannite phase predominates, varying in proportion from about 65% to over 90%, and increases with the time and temperature of heating. Occasional crystals of cuprite occur within the crednerite in the hausmannite-rich tablets. The hausmannite phase probably approaches  $\text{CuMn}_2\text{O}_4$  in composition, and has an x-ray powder pattern similar to  $\text{MnMn}_2\text{O}_4$ , differing slightly in the spacing of the lines which correspond in position to a tetragonal cell of dimensions  $a=5.75$ ,  $c=9.36$  Å.

The products of heating for 3 hours at 1135° C. a series of tablets with

TABLE 4. SYNTHETIC CREDNERITE—COMPOSITION AND CELL CONTENTS

	1	2	3	4	5	6	
CuO	64.71	52.1	48.57	51.2	50.2	Cu	1.73
MnO	33.56	42.7	46.50	44.2	44.8	Mn	1.73
O	1.73	5.2	4.93	4.6	5.0	O	4.33
Total	100.00	100.0	100.00	100.0	100.0	Density	5.00

1. Analysis by G. C. Carlos.
2. The same, after deducing 21.3%  $\text{Cu}_2\text{O}$  and recalculation to 100%.
3. Analysis by G. C. Carlos.
4. The same, after deducing 15%  $\text{CuMn}_2\text{O}_4$  and recalculation to 100%.
5.  $\text{Cu}_2\text{Mn}_2\text{O}_5$ .
6. Unit cell contents for composition  $\text{Cu}_2\text{Mn}_2\text{O}_5$ .

Cu:Mn atomic ratios greater than 1:2 include two in which crednerite predominates. One of these consists in polished section of tabular crystals of crednerite up to 0.3 mm. across, with interstitial cuprite. Micrometric analysis determined the amount of cuprite as 18% by volume. The other contains tabular crystals of crednerite about 0.03 mm. across, together with 15% by volume of the hausmannite phase. The chemical analyses of portion of these tablets are given in Table 4, (1) and (3), together with the composition of the crednerite, obtained by deducing 21.3% by weight of cuprite, (2), and 15% by weight of  $\text{CuMn}_2\text{O}_4$ , (4). In each analysis the oxygen content is taken as the difference of  $\text{CuO} + \text{MnO}$  from 100.00.

The analyses show that the composition of the synthetic crednerite is close to  $\text{Cu}_2\text{Mn}_2\text{O}_5$ , in contrast to the composition  $\text{CuMn}_2\text{O}_4$  given by the analysis of the Mendip Hills material. The analyses of Friedrichroda crednerite with the highest copper content are however more comparable with the synthetic product.

The x-ray powder pattern of synthetic crednerite is similar to that of the Mendip Hills crednerite, Table 1. The small differences in spacing of some lines indicate the  $a$  dimension of the former is 1% smaller. The unit cell content of crednerite of composition  $\text{Cu}_2\text{Mn}_2\text{O}_5$ , calculated for a cell volume 1% smaller than that of the natural material, and the measured density of 5.00 gm./cc. is given in Table 4.

The cell contents are again quite irrational, and in oxygen content lower than the Mendip Hills crystals. For the composition  $\text{Cu}_2\text{Mn}_2\text{O}_5$  the smallest multiple of the monoclinic cell which contains a nearly rational number of atoms is one of thrice the volume. However, there is no evidence in the x-ray single crystal photographs for tripling of any of the unit cell dimensions.

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## THE OCCURRENCE OF JAROSITE IN UNDERCLAYS\*

CHARLOTTE M. WARSHAW, *Gulf Research & Development Company,  
Pittsburgh, Pennsylvania.*

### ABSTRACT

Members of the jarosite family have been found intimately mixed with the clay minerals in underclays from southwestern Pennsylvania. The composition of these basic ferric sulfates has been determined by a combination of chemical and x-ray diffraction analyses. The powder patterns of these jarosites and of similar material from glauconitic sediments have been compared with indexed patterns of jarosites obtained from the U. S. National Museum.

### INTRODUCTION

Jarosite,  $\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6$ , has been identified in underclays from the southwestern part of Pennsylvania. This mineral occurs most commonly in the oxidized zones of ore deposits, but it has frequently been reported in sediments. Hutton (1950) has given representative examples of the variety of occurrences of jarosite. Briggs (1951) has shown that siltstones, glauconitic sandstones and diatomaceous shales in the Tertiary of California contain jarosite which has formed as a result of the oxidation of pyrite. Thus, the finding of jarosite in underclays which have a fairly high iron content is not surprising.

Most of the jarosite in underclays is intimately mixed with the clay minerals but some occurs as earthy yellowish patches resembling limonite. It was first noted during clay mineral studies on the supercentrifuged fractions, the equivalent spherical diameter being less than two microns. The differential thermal analysis (DTA) curve of the  $<0.3\mu$  fraction suggested the presence of a fair amount of lepidocrocite, but this was doubted since the samples were cream-colored. An x-ray diffraction pattern of the same material contained lines not attributable to the clays. Positive identification of the jarosite, which was too fine-grained for optical examination, was obtained by combining chemical and x-ray diffraction analyses.

The same mineral was also found in some glauconitic sandstones from Venezuela. It was concentrated in the  $<0.5\mu$  fractions of the clay minerals from the sandstones and seemed to be most plentiful in specimens which had yellowish material cementing the quartz and glauconitic grains. The yellow material was too fine-grained for optical identification and was studied concurrently with the underclays.

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## SOURCES OF SAMPLES

One underclay was from the Conemaugh formation in a road cut near the Youghiogheny Reservoir. A fresh sample of underclay was obtained from beneath the Upper Freeport coal in a mine at Creighton, Pennsylvania.

The glauconitic samples are from the upper member of the Cretaceous Temblador formation in the Greater Oficina area of Venezuela described by Hedberg et al. (1947). One is a well core sample from Tigre-1; the other is from Yopales-1.

## COMPOSITION OF UNDERCLAYS

The underclays from Pennsylvania consist of a mixture of disordered types of illite and kaolinite plus some quartz. In addition to the insoluble sulfate jarosite, the samples contain soluble sulfates of calcium and aluminum and perhaps free sulfuric acid. The soluble compounds were removed, and the clay was dispersed with a sodium oxalate solution. While only a minor amount of jarosite was detected by *x*-ray diffraction in the crude material, this mineral was so greatly concentrated in some of the supercentrifuged fractions that the clay minerals could not be identified before the removal of the sulfate with hydrochloric acid.

## X-RAY STUDIES AND THE IDENTIFICATION OF JAROSITE

The mineral which was later identified as jarosite gave a very distinctive *x*-ray diffraction pattern which agreed fairly well with the *ASTM*, pattern for coquimbite, but also showed some similarity to the *ASTM* patterns for jarosite and cyprusite, both basic ferric sulfates. When a jarosite-rich fraction was treated with hydrochloric acid and the residue *x*-rayed, it was found that the sulfate mineral had been dissolved, and the acid solution showed strong positive tests for ferric iron and sulfate. It was concluded that the mineral was more likely a basic ferric sulfate related to jarosite than the normal ferric sulfate coquimbite, since it was soluble in dilute acid but not in water. Proof that the sulfate was not coquimbite was obtained by *x*-raying two authentic samples of coquimbite donated by the U. S. National Museum. The diffraction pattern obtained from these samples was entirely different from the one given in the *ASTM* file for this mineral and likewise from the one obtained from the underclays. Moreover, the *DTA* curve of coquimbite differs greatly from the thermograms of the underclay samples, which agreed, at least below 600° C., with the *DTA* curves of jarosite given by Kulp and Adler (1950).

A fraction which contained a large amount of the ferric sulfate was treated with dilute hydrochloric acid, and a quantitative chemical analy-

sis was made of the solution. The ratio of ferric iron to sulfur trioxide was about the same as for jarosite, but the total alkalis were lower. Moreover, the sodium may have come entirely from the sodium-saturated clay. This analysis, No. 1 in Table 3, suggested that the mineral might be potassium-deficient jarosite. Therefore, samples of jarosite and karphosiderite, which is an alkali-free member of the jarosite family, were obtained from the U. S. National Museum for comparison.

TABLE 1. POWDER DIFFRACTION PATTERNS OF JAROSITES

(hkl)*	A			B			C		D		E	
	I	d(obs.)	d(calc.)	I	d(obs.)	d(calc.)	I	d(obs.)	I	d(obs.)	I	d(obs.)
101	3	5.94	5.93	3	5.93	5.93	1	5.95	1	5.94	1	5.95
003	2	5.74	5.74	3	5.56	5.56	1	5.73	2	5.72	1	5.65
										5.59		
012	4	5.09	5.09	4	5.05	5.05	2	5.10	3	5.10	2	5.10
110	1	3.65	3.65	1	3.66	3.66						
104	0		3.56	1	3.48	3.48						
021	6	3.11	3.11	8	3.11	3.11	2	3.120	4	3.115	3	3.115
113	10	3.08	3.08	8	3.06	3.06	3	3.085		3.080	3	3.075
									4	3.065		
202	1	2.97	2.964	2	2.96	2.963						
006	2	2.870	2.870	2	2.778	2.780	1	2.865	1	2.860	1	2.820
024	3	2.547	2.546	2	2.526	2.523					1	2.540
107	5	2.292	2.292	3	2.228	2.230	1	2.288	2	2.283		
									1	2.240		
									1	2.231		
033	5	1.978	1.976	4	1.975	1.975						
303												
027	2	1.941	1.940	1	1.905	1.905						
009	1	1.913	1.913	0		1.853						
220	5	1.823	1.823	4	1.830	1.830						
	several weak lines											
226	3	1.539	1.538	1	1.529	1.529						
02.10	3	1.512	1.512			1.476						
404	1	1.484	1.482	1	1.478	1.482						

NOTE:  $\lambda(\text{CuK}\alpha) = 1.5418 \text{ \AA}$ .

\* Hexagonal indices.

(A) Jarosite (U.S.N.M.—R6299).

(B) Karphosiderite (U.S.N.M.—R6266).

(C) Sulfate, Conemaugh underclay.

(D) Sulfate, Upper Freeport underclay.

(E) Sulfate, glauconitic sandstone.

The samples donated by the National Museum were portions of those which had been examined by Hendricks (1937) to determine the structure of jarosite. Jarosite (U.S.N.M.—R6299) from Meadow Valley Mine, Pioche, Nevada, and karphosiderite (U.S.N.M.—R6266) from Greenland were x-rayed in aluminum holders with the Norelco Wide Range Diffractometer using filtered copper radiation. Samples prepared by sedimenting the powdered minerals on glass slides were also x-rayed to determine whether these samples showed preferred orientation. Because of

its basal cleavage, the jarosite showed this effect, but the karpfiosiderite did not. Therefore, the samples were also *x*-rayed in powder cameras of 57.3 mm. radius in order to use a method of sample preparation which would give much less preferred orientation and permit better estimates of intensities. The *d* spacings greater than 1.48 Å were determined from diffractometer traces run at  $1/4^\circ 2\theta$  per minute. The powder patterns were indexed and the cell dimensions determined. These data are given in Tables 1 and 2.

The diffraction patterns of all the fractions from the underclay and glauconitic samples which contained appreciable amounts of basic ferric sulfate were examined to determine any minor differences which might be consistent for any one locality. Differences between fractions of the same samples were negligible. The major contamination was a mixture of kaolinite and illite in some fractions and quartz in others. Because of interfering diffraction lines from these impurities, the complete jarosite pattern could not be determined from any one fraction. The most en-

TABLE 2. LATTICE DIMENSIONS OF JAROSITES

Name	Formula	$a_0$	$c_0$	$c/a$
Jarosite	$\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6$	7.29	17.22	2.36
Jarosite*	$\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6$	7.22	17.03	2.36
$3\text{Fe}_2\text{O}_3 \cdot 4\text{SO}_3 \cdot 9\text{H}_2\text{O}^*$	$(\text{H}_2\text{O})\text{Fe}_3(\text{SO}_4)_2(\text{OH})_6(\text{H}_2\text{O})$	7.18	16.93	2.36
Natrojarosite*	$\text{NaFe}_3(\text{SO}_4)_2(\text{OH})_6$	7.20	16.33	2.27
Karphosiderite	$\text{Fe}_3(\text{SO}_4)_2(\text{OH})_6(\text{H}_2\text{O})$	7.32	16.68	2.28

NOTE: Dimensions of hexagonal cell are given in table;  $\lambda(\text{CuK}\alpha) = 1.5418 \text{ \AA}$ . Those marked \* are from Hendricks (1937) and have been converted to Å.

Rhombohedral cell dimensions for powder patterns given in Table 1 are: jarosite,  $a_{rh} = 7.12$ ,  $\alpha = 61^\circ 35'$ ; karphosiderite,  $a_{rh} = 6.98$ ,  $\alpha = 63^\circ 15'$ .

riched fractions from each locality were *x*-rayed with the diffractometer at slow speed in order to determine accurate *d* spacings, given in Table 1. Like karphosiderite, none of these samples showed preferred orientation. The following in particular should be compared: the spacings of the (003), (006), and (107) reflections and the spread between the (021) and (113) reflections and their relative intensities.

The Conemaugh material gave the jarosite diffraction pattern while the Upper Freeport material appears to be jarosite mixed with a smaller amount of karphosiderite. The pattern of the sulfate from the glauconitic samples is intermediate between jarosite and karphosiderite, which suggests solid solution. The resolution observed in the pattern of the Upper Freeport material shows that it is possible to detect a mechanical mixture of jarosites.

The unit cell dimensions of jarosite (Table 2) do not agree with those determined by Hendricks (1937) on a single crystal, which may be due to variations among single crystals of jarosite. Such variation was suggested by Hendricks in order to explain the discrepancy that he found between the  $x$ -ray and morphological axial ratios. Hendricks' studies indicate that  $c_0$  is more affected than  $a_0$  by substitutions in the potassium position. Karpfiosiderite, in which this position is vacant, has a very much smaller  $c_0$  than jarosite, while the  $a_0$  dimensions are almost the same. Natro-jarosite is also characterized by a smaller  $c_0$ . When water molecules fill the potassium position as in synthetic  $3\text{Fe}_2\text{O}_3 \cdot 4\text{SO}_3 \cdot 9\text{H}_2\text{O}$ , the axial ratio is very similar to that of jarosite.

#### CHEMICAL STUDIES OF BASIC FERRIC SULFATES IN SEDIMENTS

The  $x$ -rayed fractions containing the water-insoluble sulfates were given a mild hydrochloric acid treatment, the results being shown in

TABLE 3. CHEMICAL ANALYSES OF SAMPLES CONTAINING JAROSITES

	Weight Per Cent			Molecular Equivalents		
Jarosite—Theoretical						
K <sub>2</sub> O	9.41			1.00		
Fe <sub>2</sub> O <sub>3</sub>	47.83			3.00		
SO <sub>3</sub>	31.97			4.00		
No. 1—Upper Freeport underclay <0.4 $\mu$ Fraction						
Na <sub>2</sub> O	1.11			0.39	} 0.77	
K <sub>2</sub> O	1.61			0.38		
Fe <sub>2</sub> O <sub>3</sub>	24.54			3.31		
SO <sub>3</sub>	14.93			4.00		
	Na <sub>2</sub> O	K <sub>2</sub> O	SO <sub>3</sub>	Na <sub>2</sub> O	K <sub>2</sub> O	Total Alk.
No. 2—Upper Freeport underclay 2-0.4 $\mu$ fraction	1.44	1.20	11.50	0.65	0.36	1.01
No. 3—Conemaugh underclay	1.08	0.72	4.19	1.33	0.58	1.91
No. 4—Glaucconitic sandstone	0.9	0.64	4.02	1.16	0.54	1.70

Table 3. All the sulfate was assumed to be present as jarosite. Thus, the molecular equivalents of the alkalis are based on a figure of 4.00 for the molecular equivalent of SO<sub>3</sub>. The sulfates in the sediments are deficient in potassium. The highest percentage of sodium was found in a sample which contained more jarosite than some of the others. Thus, fractions which appear to give a karpfiosiderite  $x$ -ray pattern may ac-

tually contain natrojarosite. It is apparent from the lattice dimensions in Table 2 that natrojarosite must have a diffraction pattern more similar to karpfoserite than to jarosite.

Soda and water determinations are of no value in determining the compositions of the jarosites in these samples because of the admixed clay minerals. The best that can be done is the estimation of probable compositions by considering both the  $x$ -ray diffraction patterns and chemical analyses for sulfate and potash.

The Upper Freeport underclay, which contains much less  $K_2O$  than the other samples, gives the diffraction pattern of a mixture of jarosite minerals. It contains relatively pure jarosite and either karpfoserite or natrojarosite. If the latter is present, it must contain some vacant alkali positions in the structure since the total alkalis are lower than the theoretical amount. The jarosites from the Conemaugh underclay and the glauconitic sandstone, though very similar in chemical composition, give different  $x$ -ray patterns. Since the sulfate from the Conemaugh underclay gives a pattern of practically pure jarosite, many of the alkali positions may be filled with water molecules rather than sodium ions. The jarosite associated with the Venezuelan glauconite is a solid solution with only half of the alkali positions filled by potassium. It cannot be stated definitely whether the others are vacant or contain sodium, but the latter is probable since this particular sample was not appreciably contaminated with sodium saturated clay.

#### THERMAL STUDIES OF BASIC FERRIC SULFATES IN SEDIMENTS

Differential thermal analysis, carried out with apparatus described by McConnell and Earley (1951), first indicated that an unusual constituent

TABLE 4. THERMAL DATA FOR JAROSITES

	Endothermic Peaks		Exothermic Peaks
Jarosite (Kulp and Adler—1950)	470° C.	800° C.	590° C.
Upper Freeport underclay	430° C.	790° C.	490° C.
Conemaugh underclay	425° C.	770° C. ?	470° C.
Glauconitic sandstone	390° C.	—	425° C. ?

was present in these underclays. Before it was identified as jarosite, combined  $x$ -ray diffraction and differential thermal studies (Table 4) were carried out on fractions of the Conemaugh underclay and the glauconitic sandstones. The  $x$ -ray patterns remained unchanged up to the start of the first endothermic reaction, during which the color changes from light gray or cream to brown and the  $x$ -ray diffraction lines of the



sulfate mineral disappear. After the material has been heated for a longer time or at a higher temperature, it takes on a red color and gives the pattern of hematite. Most of the samples in Table 4 show no definite reaction attributable to jarosite at about 800° C. Apparently a large minimum percentage of jarosite is necessary for developing the characteristic thermogram given by Kulp and Adler (1950). Thus a small amount of jarosite in a sediment may be confused with lepidocrocite,  $\text{FeO}(\text{OH})$ , if the differential thermal method is used for identification. The peak temperatures found in this study differ from those of Kulp and Adler probably because of their faster heating rate (12.5° C. instead of 10° C. per minute) and the impurity of the jarosites concentrated from the sediments.

#### FORMATION OF JAROSITE IN SEDIMENTS

Merwin and Psonjak (1937) have discussed the chemistry of sulfate formation from the oxidation of pyrite in ore deposits based on their equilibrium studies in the system  $\text{Fe}_2\text{O}_3\text{-SO}_3\text{-H}_2\text{O}$  (Posnjak and Merwin, 1922). Complete oxidation of pyrite in the presence of sufficient water should yield the acid ferric sulfate rhomboclase,  $\text{HFe}(\text{SO}_4)_2 \cdot 4\text{H}_2\text{O}$ , but the iron oxidizes more slowly than the sulfur and a mixture of ferrous and ferric sulfates and sulfuric acid results at first. The acid reacts with other minerals in the rock thus lowering the acidity of the solution. Alkalies and alkaline earths are easily introduced into the solution in this manner, especially if clays with exchangeable cations are present. When considerable dilution takes place, for example, when leaching and oxidation occur together near the surface of the ground, basic ferric sulfates may be precipitated. The jarosite type of basic sulfate occupies a large field below 170° C. in the central portion of the phase equilibrium diagram given by Posnjak and Merwin. The substitution of different alkalies in the jarosite structure should not greatly affect the general conditions of equilibrium. With considerable or continued dilution, the final product of the leaching and oxidation of pyritic rocks is goethite; however, once jarosites are precipitated, they are not readily dissolved.

The process described above should also apply under certain conditions to the oxidation of pyrite in sediments. Pyrite is very common in glauconitic sediments, coal beds, and other formations in which a reducing environment has been maintained. The occurrence of sulfates in any of these formations would be indicative of a considerable change in environment. Oxidation could occur more readily in sandstones and underclays than in sediments which contain fairly large amounts of organic matter. Dilution of the resulting acid solutions, which is also necessary for the deposition of basic ferric sulfates, could take place more readily

in porous sandstones than in clays or shales. On the other hand, sulfates might be completely leached out of sandstones and deposited elsewhere.

In most of the glauconitic samples, the jarosite was associated with kaolinite and relatively soluble calcium sulfate in the interstices between grains of quartz and glauconite. In some cases, it was concentrated in small lenses (a few inches in diameter) with kaolinite, fine-grained quartz and soluble calcium and aluminum sulfates which give an acid reaction with water. These lenses may have formed when percolating solutions became loaded with fine-grained material, clogged pores and deposited the mass of sulfates and clay in such a manner that further leaching could not occur. It is concluded that the jarosite found in the glauconitic sediments from Venezuela is merely a precipitate from the oxidation of pyrite and the dilution of the resulting solutions. The unaltered glauconite shows no evidence of replacement by jarosite like that observed by Briggs (1951) in California sediments.

Dilution probably is not as important in the deposition of jarosites in underclays as in sandstones. In fact, these underclays appear to contain free sulfuric acid and would be expected to contain soluble as well as insoluble sulfates. It is interesting, therefore, that soluble ferric iron was associated with the aluminum and calcium sulfates in the Upper Freeport underclay but not in the Conemaugh underclay, which had been exposed in a road cut.

#### SUMMARY

Members of the jarosite family of basic ferric sulfates have been found in underclays from the southwestern part of Pennsylvania. Because the particle size of the sulfates was less than one micron, they could not be separated mechanically from the clay minerals nor could they be identified optically. The probable composition was determined by a combination of chemical and  $x$ -ray diffraction analyses. Differential thermal analyses were also made.

$X$ -ray diffraction patterns of powdered samples of jarosite and karpfoidite from the U. S. National Museum facilitated the identification of the basic ferric sulfates in the underclays. The powder patterns were indexed and lattice dimensions determined. The *ASTM*  $x$ -ray diffraction pattern for coquimbite was found to be a pattern of a member of the jarosite family.

The  $x$ -ray and chemical analyses indicated that jarosite is present in the underclays but that some of the potassium positions are probably filled with water molecules while others may contain sodium. One underclay sample, with less potassium in combination with the sulfate, contained a mixture of jarosite with natrojarosite or karpfoidite. A basic

ferric sulfate in glauconitic sandstones from Venezuela was found to be a solid solution of jarosite and natrojarosite with a composition closer to the former member.

The formation of jarosites in pyritic sediments during concurrent oxidation and leaching processes is consistent with the phase equilibria studies in the system  $\text{Fe}_2\text{O}_3\text{-SO}_3\text{-H}_2\text{O}$  carried out by Posnjak and Merwin (1922). Both the iron and the sulfur are derived essentially from the pyrite, while the alkalis are derived from the other minerals of the sediments, particularly illite, by acid attack.

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## MEMORIAL OF ALTON GABRIEL

T. N. McVAY, *U. S. Bureau of Mines, Tuscaloosa, Alabama.*

The writer worked in Alton Gabriel's office the week preceding his death. He appeared to be in good health, was jovial, and was making plans for the future. However, on Saturday, October 23, 1954, he suffered a fatal heart attack.

Alton Gabriel was born in Portland, Oregon, September 2, 1902, and was the son of Albert Gabriel and Bertha Rothlesberger Gabriel, who were early settlers of Oregon. He attended public schools in Portland and later graduated from the University of Oregon in 1925 with the degree of Bachelor of Science in Chemistry. He received his Master of Science degree in Chemistry in 1927 and the subject of his thesis was "Use of aluminum oxide in the refining of kerosene distillates." After graduation he was instructor in analytical chemistry for one year at the University of Oregon. He then went to Cornell University where he was a graduate assistant and received the Ph.D. in Chemistry in 1930. The subject of his dissertation was "Geochemical data of germanium." Thus, he anticipated the great current interest in germanium by almost 25 years. After graduation he was research chemist for Roessler and Hasschlacher. About this time Everett P. Partridge, of the Bureau of Mines, was interested in having a chemist-spectrographer make mineralogical studies of the New Mexico salt minerals, and persuaded Alton to join the staff of the Bureau of Mines at New Brunswick, New Jersey. He rose rapidly and steadily in the Government service to the position of Assistant Chief of the Minerals Division, Metallurgical Branch, Washington, D. C. In January 1950 he was made Assistant Director of Region VII, with headquarters at Norris, Tenn., and after the death of Hewitt Wilson, he assumed the position of Acting Director of the region. However, Alton's major interest was in scientific studies rather than administrative duties, and in February 1953 he transferred to the Southern Experiment Station of the Bureau of Mines, Tuscaloosa, Ala., where he was Chief of the Ores and Metals Branch, Region VII, and was busily engaged in mineralogical studies at the time of his death.

He was a member of the American Chemical Society, Fellow of the Mineralogical Society of America, Society of the Sigma Xi, and Delta Upsilon. His bibliography, while not lengthy, is in no way a criterion of the large amount of important work accomplished during his professional career, although his list of publications does indicate his wide scientific interests. Alton became one of the most competent and versatile chemical microscopists and petrographers of our time. He was able to correlate the use of the microscope, x-ray, spectrograph and differential thermal



analysis for the solution of many difficult mineralogical problems. He is to be remembered as one of the first to recognize the importance of the spectrographic analysis in connection with the study of minerals, and this probably stemmed from his early training under Jacob Papish and Chamot and Mason at Cornell University. He also adapted petrographic



ALTON GABRIEL  
1902-1954

techniques to the study of mineral concentrates so that the ore dressers have a positive and rapid evaluation of their progress in the beneficiation of complicated ores. Consequently, during these years he exerted a profound and lasting influence on mineralogists and petrologists who apply their techniques to the solution of mining, metallurgical, mineralogical, and ceramic problems of the Bureau of Mines, and the methods he devel-



oped have also been widely applied in university and industrial laboratories. He was particularly effective in working with beginning scientists in the Bureau, and his advice was perhaps a major factor in the advancement made by many of them in this field. He took particular interest in seeing that a number of young and promising boys were given part-time employment to help defray their college expenses. In at least one instance the Gabriels used personal funds in aiding a student through medical school.

Alton Gabriel married Lola Jacobs August 17, 1932, and there are no living children.

He will long be missed by all who knew and worked with him, and this quotation from the *Analects of Confucius* describes him well:

The Master was entirely free from  
four things: prejudice, foregone  
conclusions, obstinacy, and egoism.

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## MEMORIAL OF JOHN PUTNAM MARBLE

WILLIAM F. FOSHAG, *U. S. National Museum, Washington, D. C.*

John Putnam Marble was born in Worcester, Massachusetts, May 30, 1897, and died suddenly in Washington, D. C., June 6, 1955. His scientific career was that of an amateur, one who cultivates his science from taste, without pursuing it for gain. As such, Marble was willing, and able, to undertake unrewarding tasks, necessary to the advancement of his chosen field, and which others more professionally inclined, neglected as not being sufficiently flamboyant. His strong sense of duty to science and to humanity undoubtedly stemmed in large part from his Quaker heritage. He traced his ancestry to the Pilgrim fathers; and counted, I understand, among his collateral forebears Peregrin Smith, the child born upon the Mayflower. He showed an early love for the out-of-doors, which led him to an appreciation of geology; the family dry-saltery business, on the other hand, developed an interest in chemistry. These two interests he later combined successfully into a career in geochemistry.

After attending Worcester Classical High School, he entered Williams College, where he attained Phi Beta Kappa in his junior year, and graduated *cum laude* in 1918, and as co-recipient of the John Sabin Adriance Prize in Chemistry.

He entered the Chemical Warfare Service of the Army in 1918, serving in the Research and Control Laboratory, Edgewood Arsenal, receiving his discharge in early 1919. After a brief interval as part-time special student at Clark University, he entered the family dry-saltery business, J. Russel Marble and Company.

In 1921 he married Adelaide Holme Maghee. Marble was not particularly interested in a career as merchant. With the urging of his wife, he decided to retire as drysalter, and become a scientist. He began advanced work at Harvard University in 1926, received his M.A. in chemistry in 1928, and his Ph.D. in analytical chemistry under G. P. Baxter in 1932.

His doctoral thesis at Harvard was a determination of the atomic weight of phosphorus, an exercise that required the preparation of his reagents in the purest possible form. This preoccupation with purity of preparations was an important requirement in his later researches in determining the absolute age of minerals.

In 1931 he began his association with the Committee for the Determination of Geologic Time, of the National Research Council, A. C. Lane, Chairman, as Research Associate. He began a program of geologic age determinations, based upon the lead-uranium and lead-thorium ratios of radioactive minerals. He also began the compilations of the

bibliography of the literature relating to the measurement of geologic time. In recognition of his devotion to the work of the committee he was made vice-chairman in 1936. Upon the death of its chairman Lane in 1946, he was named chairman. For a number of years Marble was essentially the Committee on the Determination of Geological Time; its annual report, widely recognized as one of the most valuable and popular of the National Research Council's reports, became almost completely his work.

His work with the Committee for the Determination of Geological Time may be divided into two categories: (1) original researches on absolute geological age measurements; (2) collation of the published information relating to geochemistry, in the form of an annual summary and comprehensive annotated bibliographies.

When Marble began his researches upon the absolute determination of geological time, the only practical method was the laborious analysis of lead, uranium and thorium. Uraninite or pitchblende was the preferred material to work with, but this mineral was then considered a rarity, and restricted in geological environment. Allanite is more widespread but contains only traces of uranium and lead. Working, however, with considerable quantities of sample, and exercising care and patience, significant results could often be obtained with this mineral. Through months of tedious and meticulous chemical manipulation, the geological age of a number of such specimens were obtained.

The annual annotated bibliographies on geological time constituted a tremendous task for one person to complete singlehandedly. The world's literature was combed systematically, significant articles in many languages were read, evaluated and abstracted. The completeness of the published bibliographies, which do not include, of course, the numerous articles perused but rejected as without pertinence, attests to Marble's assiduous labor in their preparation. The last bibliography, covering the year 1953-1954, contains 635 entries.

In addition to these important contributions to the work of the committee, Marble maintained contact with other workers in this field throughout the world, arranging for interchange of ideas, of materials for study, or in other desirable ways. Sometimes the results of these contacts appeared as appendices to the Committee's report; translations of significant articles not readily available to American workers, or the personal reports of foreign collaborators.

Marble also continued his interest in his early work on the atomic weights of the elements, and spent the summers from 1931 through 1942 at Harvard working on problems in this field. One of these involved the determination of the atomic weight of scandium. In order to obtain sufficient material for this project he personally arranged for the reopening



JOHN PUTNAM MARBLE  
1897-1955

of the thortveitite-bearing pegmatite at Iveland, Norway.

One must finally mention his interest in meteoritics, which manifested itself in a description of the Osseo, Canada, octahedrite. He was also a member of the International Committee for the Study of Meteorites, organized during the 18th International Geological Congress in 1948.



In 1935 Marble became associated unofficially with the Smithsonian Institution. In 1948 he was named Associate in Mineralogy, in recognition of his interest and assistance in the work of this Institution. He was appointed its official delegate to the 17th International Congress, U.S.S.R., the 18th, England, and the 19th, Algiers. He was secretary general of the American Geophysical Union, and attended the Tenth General Assembly of the International Union of Geodesy and Geophysics at Rome in 1954. In all these he contributed his own expenses. He was also secretary-treasurer of the American Geological Union, and a councillor of the American Association for the Advancement of Science. During World War II he served as technical aid and special assistant of the National Research Committee of the Office of Scientific Research and Development.

Marble was a fellow or member of a number of scientific societies, including the Mineralogical Society of America, Geological Society of America, American Geophysical Union, American Chemical Society, Electrochemical Society, American Meteoritical Society, American Association for the Advancement of Science, the Washington Academy of Sciences, and the New York Academy of Science.

Marble's work was a labor of love. He might have pursued his science desultorily in the manner of the usual dilettante. Instead he spent long hours daily in his scientific pursuits and took his responsibilities with inordinate seriousness. His independent position freed him of the common preoccupations of most scientists, and lent him the air of the old-school intellectual. The Spanish would class him as an "educated" man, which involves not only learning, but broad cultural attainments. His home life was gracious and dignified. His interests were broad and included not only his science, but art and literature as well. He was particularly interested in education, and in his capacity of Trustee of the Sidwell's Friends School, he studied the advances of progressive education as practiced in Europe.

Probably his most obvious attribute was unselfishness. Although he often grouched good naturedly, he never refused a task imposed upon him.

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## MEMORIAL OF CHARLES PALACHE

CLIFFORD FRONDEL, *Department of Mineralogy, Harvard University,  
Cambridge, Massachusetts.*

Charles Palache was born July 18, 1869, and died December 5, 1954, in the eighty-sixth year of his age. One of the most eminent crystallographers and mineralogists of the world, he lived in a period of revolutionary developments in mineralogical science.

Palache's ancestors belonged to a group of Sephardic Jews who at the end of the 15th Century were exiled from Portugal to Holland. Much later one family migrated to Jamaica where Charles Palache's grandfather, John, headed a plantation. For political reasons he abandoned that home in 1834, put his wife and three daughters on a ship sailing for New York, but died before he could follow them in the next boat. Three months later Palache's father, James, was born in New York City. Lured by reports of gold in California, James left his home at the age of fifteen to serve as cabin boy on a schooner rounding Cape Horn. He landed in San Francisco in 1849. There, established as a merchant, he married Helen D. Whitney, who had traveled from her home in Green Bay, Wisconsin, in a caravan of seven covered wagons. Their son, Charles Palache, was a sensitive boy who at an early age evidenced an intense interest in nature and collected objects of natural history. Palache graduated from Berkeley High School, and entered the University of California in 1887. He elected the four year course in mining, since in its content there was more natural history than in any other, and graduated at the top of his class. Andrew C. Lawson was appointed Professor of Geology in his senior year, and Palache returned the following year to assist Lawson in mineralogy and to study for the doctorate, which he received in 1894. Lawson, himself at the start of a long and distinguished career, was a stimulating teacher and it is to him that Palache credited the inspiration that took him from a career in mining into mineralogy. At first Palache's interests were in field geology and petrography, and with Lawson he did the field work for the first geologic maps of the San Francisco Peninsula and the Berkeley area.

In 1894, Palache left for a year of study abroad, first to work under Ferdinand Zirkel at Leipzig, where T. C. Walker and Bundjiro Koto were fellow students, and then to study with Paul Groth and Ernst Weinschenk at Munich. Other American students working in Groth's laboratory at the time were T. A. Jaggar, A. B. Peck and A. S. Eakle. The winter in Munich was a happy and busy one; and it is recorded in Palache's journal that "the museums were good, the theatre excellent

and there was opera all winter costing students next to nothing." A turning point in his life came the following spring, when he visited Heidelberg. Here he took courses in petrography under Harry Rosenbusch and Alfred Osann, and was introduced to morphological crystallography by Victor Goldschmidt. Palache threw himself with enthusiasm into the study of crystals, and laid the foundation for the work he pursued so vigorously for the next fifty-five years.

Palache returned to California in the fall of 1895, and in December a letter came from John E. Wolff offering him a small job as his assistant



Professor Charles Palache working at the Goldschmidt two-circle goniometer. Photographed in 1936.

at Harvard. Wolff, Professor in the then newly organized Department of Mineralogy, and Curator of the Mineralogical Museum, succeeding Josiah P. Cooke, was one of a group of Harvard geologists that included William Morris Davis, Nathaniel S. Shaler and Josiah D. Whitney. Wolff's interests were primarily in petrography and in the year after his retirement in 1922 the instruction and research in this field was taken over by Esper S. Larsen, Jr., who with Palache constituted the Department for many years. Palache was named Assistant Professor of Mineralogy in 1902, Professor in 1910, and Professor Emeritus after his retirement in 1941.



## MORPHOLOGICAL STUDIES

In his chief field of work, morphological crystallography, Palache brought himself and the Department of Mineralogy to a pre-eminent position in research on the external geometry of crystals. There is scarcely a crystallized mineral that he did not investigate. He introduced the first Goldschmidt two-circle goniometer into the United States, in 1896, and elaborated this method in a series of papers that with later amplifications by his students are standard references. The present general use in America of two-circle goniometric methods in the characterization of crystallized substances derives largely from his work.

Palache's publications deal chiefly with systematics and descriptive matters, and it is through the work of his students that we see the keen and stimulating interest he had in the genetic and interpretive aspects of mineralogy. His published papers, over 150 in number, include classical studies of the morphology of calcite, azurite, the gold tellurides, the lead oxyhalides, and definitive investigations of numerous less common minerals. His studies of calcite culminated in 1943 in a critical review of the morphology of this species that surpasses the early works of Bournon, Zippe and Irby. His study of the tantalizing mineral calaverite, done in cooperation with Martin Peacock and Victor Goldschmidt, is a masterpiece of the art of crystal measurement and projection. Peacock began the study of crystallography under Palache in 1926, and returned as Research Assistant to him over the period 1932-1937. A brilliant man, with a gift for conciseness in style and rigor in presentation, he gave to Palache a kindly affection and shared with him both a love of music and skill at the carpentry bench.

Palache's published studies are only a small part of the crystallographic work that he accomplished. In one project the available morphological data for all minerals was critically re-examined, recalculated in Goldschmidt's scheme of presentation, and tabulated on filing cards. This modern version of Goldschmidt's *Winkel Tabellen* was not brought to full completion, however, partly from the realization that the new x-ray methods of study would in many instances necessitate a description different from that based on morphological grounds. There was further the growing realization, emphasized in the Harvard laboratory by Harry Berman, that the features of minerals were not isolated matters but should be described in context with the chemical composition and the crystal structure. Berman, the nearest to a son that Palache had, went to Harvard in 1924. He was versed in modern chemistry and physics, which Palache was not, and became a powerful stabilizing influence in the laboratory. Berman installed the first x-ray equipment at Harvard in 1933. The purely geometrical point of view of earlier years, that at times



yielded ornamentations of morphological crystallography bordering on numerology, became realistic and broadly based.

The preparation of the 7th edition of Dana's System of Mineralogy was started in 1937 under Palache's leadership. He gave close attention to the problems of organization and computation of the crystallographic data, with the help of C. W. Wolfe and Peacock, and the files of the Dana contain a large store of measurements and computations that came from his hand. Although he did not prepare any of the manuscript, his counsel and factual knowledge contributed greatly to the progress of the work.

Crystals had a deep significance to Palache. In their morphology he seems to have found almost an element of mysticism, a response to the facets of a crystal seen in glittering progression in the quietness of a goniometer room, that revealed a self-contained system of order in a random world. The resolution of this order in terms of atoms and forces did not attract him. The full impact on mineralogy of the new *x*-ray and physical techniques came midway in his career. As a scientist he welcomed and used the knowledge thus brought of the ultimate structure of crystals, yet this new understanding terminated a traditional approach to the constitution of crystalline solids, one followed in mineralogy for a century and more, and destroyed for him the wonders of a thing unknowable and long sought.

#### THE MINERALOGICAL MUSEUM

Palache's most lasting and most important contribution to the development of the Harvard Department of Mineralogy, and a great service to mineralogy in general, was in building the Mineralogical Museum to its present position as the leading research and exhibit collection of minerals in the world. When Palache first came to Cambridge, a few days before Christmas in 1895, it was to assist Wolff in arranging the mineral collection. Palache lived for a year in a small room in the University Museum where, armed with a rifle, he guarded the premises.

The collection he came to had started in 1784 and in 1895 contained about 55,000 specimens. Wolff continued as Curator until 1922, when he retired and Palache took charge both of the Department and of the Museum. The collection grew rapidly by field collecting, exchange and purchase. The great private collection of A. F. Holden, comparable in quality and extent to the collections of Roebbling and Bement, was acquired by gift in 1913. This was followed by very large funds given by A. F. Holden in 1922 and by J. E. Wolff in 1940 for the care and increase of the Museum. Much of the income from these gifts was diverted into the general funds of the University and only a small amount remained for minerals.

Beginning in 1904, Palache gradually built a definitive collection of the minerals of Franklin, New Jersey, and acquired together with a mass of other material the Hancock collection and, jointly with the U. S. National Museum, the Canfield collection. His monographic study of the mineralogy of the Franklin ores, published in 1935 by the United States Geological Survey as Professional Paper 180, is a landmark in American mineralogy. The work at Franklin was part of a lengthy association with the Geological Survey that included field studies in 1901 in the Bradshaw Mountains in Arizona and mineralogical studies in 1906 and 1919-1921 in the Lake Superior copper district.

Palache first became seriously interested in the mineralogy and paragenesis of the pegmatites of New England in 1912 although he had earlier described with C. H. Warren the pegmatite pipes of the Quincy granodiorite. In the summer of that year he collected in the pegmatites of Maine and New Hampshire, and secured the fabulous find of purple apatite at Mount Apatite near Auburn, Maine. During the next 20 years he collected, with the assistance of students and of F. A. Gonyer, extensive suites of material from pegmatites throughout New England, in part by leasing operations at important localities. This material served as the basis of important studies by himself and his students. Among these publications was Landes' well-known paper of 1925, "The paragenesis of the granite pegmatites of central Maine." Pegmatite mineralogy was strongly emphasized in Palache's course on mineral paragenesis, but he also was keenly interested in other types of mineral occurrence and it is a pity that only a small part of his store of knowledge in this field was ever published.

There were numerous other collecting activities, both at home and abroad. In 1922 he participated with R. A. Daly, F. E. Wright and G. A. Molengraaff in an expedition to South Africa, where he obtained a wealth of secondary zinc, copper and vanadium minerals. In 1924 he made a productive trip to localities in Norway and to Långban, Sweden, where the Flink collection was secured. In 1926 he went to Madrid for the XIV International Geological Congress, and to Lisbon, where he acquired the Bello collection of Portuguese minerals. Earlier, he had visited Russia for the VII International Geological Congress in 1897, and he traveled to Alaska in the Harriman Expedition of 1898. In 1935, Palache visited Vienna to purchase 900 superlative specimens from the Karabacek collection, a transaction remembered to this day in Austria. Another large acquisition was the Ahlfeld collection of Bolivian minerals, purchased jointly with the National Museum. The collection of meteorites, originally acquired from J. Lawrence Smith in 1883, was increased as opportunity offered. Palache prepared a new catalogue of this collection, and described five new meteorites.

Numerous publications stemmed from these accessions, and a vast amount of research material remains untouched. There were many indirect yields from the collection. L. C. Graton paid tribute to Palache for his essential aid in assembling the reference collection of polished sections of ore minerals in the mining geology laboratory, that formed the basis of the works of J. Murdoch, C. M. Farnham and M. N. Short in this field. Palache published joint descriptions with others of 17 new mineral species including the last two rock-forming minerals to be recognized, lawsonite and pumpellyite. Lawsonite was originally found by Palache and F. L. Ransome when as students at the University of California they went on a collecting trip to the glaucophane schists of the Tiburon Peninsula. Palache prepared a description of lawsonite in 1894 while working under Groth in Munich, and correspondence brought out that Ransome was working on the mineral in California; both had independently selected the name lawsonite, and a joint description was published in the *Zeitschrift für Kristallogie und Mineralogie*. Palacheite, named after him in 1903 by Eakle, was soon discredited as a variety of botryogen.

Palache was a connoisseur of minerals. He could evaluate the worth of a specimen as representative of a locality or type of occurrence; he knew the subtleties of crystal habit, color, association and size that distinguish a fine specimen from a good one; he was a keen judge of the factors that determine the aesthetic and scientific values. And, of course, he was a master at that virtually lost art, sight identification. Palache took a keen delight in a beautiful specimen, yet with sober deliberation he would yield any specimen to the dissecting chisel and hammer if new knowledge could be obtained. He took painstaking care in the arrangement, cataloguing and labelling of the collection to make it convenient for use by the investigator and the student. Palache's lectures were enlivened by anecdotes of his personal experiences with other mineralogists or of his visits to famous mineral localities. His students and assistants soon learned of his great knowledge and love of minerals and inevitably became imbued with his interest and spirit of research. The laboratory work was thorough and emphasized the direct examination of specimens. His successive course assistants in the earliest years were A. S. Eakle, C. E. Lord, Hoyt S. Gale, H. O. Wood, R. W. Richards, H. E. Merwin, R. E. Somers, W. G. Foye and A. Wandtke, all of whom went on to distinguished careers.

Palache played an active part in the organization and later development of the Mineralogical Society of America. The Society was first organized on December 30, 1919, in a meeting in the mineralogical lecture room at Harvard. Palache became President of the Society in 1921,

Honorary President in 1950, and was the first recipient of the Roebling Medal in 1937. In the words of Edward H. Kraus, the Roebling award was presented to "... America's foremost mineralogist, and one of the stalwarts of the Society, whose publications during a period of 40 years have covered a wide range of subjects and have contributed signally and enduringly to the advancement of our science." Palache's acceptance was a delightful account of his friendship with Colonel Roebling and of the circumstances through which he brought the Roebling endowment to the Society. Palache was loath to have the relatively large number of contributions from himself and his students and associates impose upon the limited resources of the journal of the Society. This brought the issuance of five independently financed Harvard numbers of the journal, including the Palache *Festschrift* of 1938, that supplemented the normal contributions of the Department.

The distinction of Palache's career brought him many honors. Aside from his recognition by the Mineralogical Society of America, he was a member of the National Academy of Sciences, the American Academy of Arts and Sciences, President of the Geological Society of America in 1937, and corresponding member of the Geologiska Föreningen, Stockholm. He was an Honorary Member of many societies, including the Sierra Club, the New York Academy of Science, the Mineralogical Society of Great Britain, the Royal Geological Society of Cornwall, and the Société Géologique de Belgique. In 1941 he was given an honorary LL.D. by the University of California. He was an associate editor for many years of the *Zeitschrift für Krystallographie* and of the *American Journal of Science*. Palache encouraged the efforts of the amateur mineralogists, and was voted an Honorary Member of both the Boston Mineral Club and the New York Mineralogical Society.

In his office and laboratories Professor Palache seemed to many to be stern in attitude and almost forbidding in appearance. He was not easy of approach, although an effort always was rewarding, and he rarely was familiar in his relations with students or his associates. These characteristics may have stemmed from an acute and strongly disciplined shyness and sensitivity. Certainly he was a modest and considerate person, whose kindnesses were unobtrusive. At home he was a relaxed and charming host. He derived great satisfaction from symphonic music, a taste dating to concerts at the Gewandhaus in Leipzig heard as a student. His personal reserve was easily penetrated by discussions of music or through his interest, maintained over the years, in ornithology. Palache married Helen Markham in 1898, and he is survived by three daughters. It was a pleasant privilege to visit his home or his summer place at Jaffrey, New Hampshire. Vigorous in body and mind to the end,



he died at his home near Charlottesville, Virginia, to which he had moved from Cambridge a few years before.

Such was the nature of the man, and of his work. Shortly before his death, in conversation with Reginald A. Daly, a friend and associate over many years, Palache remarked that his life had been fruitless—but this is a feeling given in great times, to great men.

\* \* \*

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## MEMORIAL OF ROBERT CHARLES WALLACE

J. E. HAWLEY, *Queen's University, Kingston, Ontario.*

Dr. Robert Charles Wallace, former Principal and Vice-Chancellor of Queen's University, and a charter member of The Mineralogical Society of America died at his home in Kingston on January 29, 1955.

Beginning his career as a demonstrator in crystallography at Aberdeen University, Wallace became successively professor of geology at the University of Manitoba, Commissioner of Mines of Northern Manitoba and of the province as a whole, and later head of two other universities, Alberta and Queen's. In his earlier years he contributed much to the science of mineralogy and geology and to the development of the mineral resources of Canada, but as an administrator and one of Canada's foremost educationalists his work set a record which has rarely been excelled and will seldom be equalled.

Dr. Wallace was born in 1881, in the Orkney Islands where he grew to manhood and himself acquired the qualities he thought so typical of the islanders, "qualities of reliability, loyalty, application to work, and love of learning . . .," a certain Scottish quality of humor, and "a natural mysticism which . . . colour(ed) all his thinking . . ."

His early education was obtained at Deerness and Kirkwall where an ingenious headmaster incited his curiosity and interest in the natural sciences and influenced his later specialization at Edinburgh under Sir Archibald Geike where he obtained his Master of Arts degree in 1901 and Bachelor of Science in 1907.

From 1907 to 1909 he pursued graduate work at Göttingen University under Professor G. Tamann, petrologist and one of the early geochemists, and completed his doctorate thesis on the "Binary systems of sodium-barium-strontium metasilicates and the three component system soda-alumina-silica."

On his return to Scotland, Wallace continued his studies at St. Andrew's University on the crystallographic relationships of indium and thallium, acting also as a demonstrator in crystallography. One can well imagine the place he might have made for himself among crystallographers and geochemists had he continued his researches in this direction. His broader interest in education as a whole was developing even at this time and is shown by early papers on the teaching of science and mathematics in the German schools.

Opportunities in Canada, however, beckoned and, in 1910, he joined the University of Manitoba as a lecturer in mineralogy and geology and within two years rose to head of the department, a position he retained until 1928. Among his early investigations as a part-time officer of the



ROBERT CHARLES WALLACE  
1881-1955

Geological Survey of Canada, were the genetic relationships of gypsum, anhydrite and salt deposits of the Silurian in Manitoba and the intriguing problem of dolomitization.

Following the successful establishment of gold mining in Northern Ontario, attention was directed to the Precambrian of Northern Manitoba. Discoveries and early development of the rich copper and zinc ores of the Mandy and Flin Flon, and of gold in Herb Lake and Pipe Lake, soon focused attention on the mineral possibilities of these areas. No better choice could have been made than Wallace's appointment, in 1918, as Commissioner of Mines for Northern Manitoba, a position he held while on leave of absence from the University, till 1921.

Much of his work in this post was of an administrative nature, supervising an area of some 178,000 square miles, and only a small part is shown by his comprehensive papers on the mineral resources of the province. Much of the credit for the development of the Flin Flon deposits, according to Dr. G. M. Brownell, must go to him, and his advice undoubtedly led to the building of the railway from The Pas to Flin Flon. It was, however, in his day to day duties, in dealing with prospectors, promoters, and mining magnates that he showed his outstanding qualities, a sympathetic understanding, sound judgment and "a business acumen frequently lacking in men of academic attainments" (Hague, R. H., 1921—*Can. Mining Journal*, 42, 749.)

As a sequel to his intimate acquaintance with this area came a major contribution (1921) in his discussion of the transfer of the natural resources of Manitoba, Saskatchewan and Alberta from the federal government to the provinces.

On his return to the University, according to Dr. J. S. DeLury, a close associate, he carried a very heavy teaching load, as well as assuming the duties of Commissioner of Mines for the entire province. In spite of this he found time to prepare several papers on both the metallic and non-metallic resources of the province and on a wide range of scientific subjects such as the characteristics of the Flin Flon ore bodies, colloidal processes in rock weathering, and the clays of Lake Agassiz.

In recognition of his standing in the Canadian mining industry, Dr. Wallace was elected successively as councillor (1919-1920), vice-president (1921-23) and president of the Canadian Institute of Mining and Metallurgy (1924), and in subsequent years was frequently called on to present his views on current problems confronting the industry.

In 1928 he was called to Alberta as president of the University and from then on his main efforts were directed toward the broader aspects of education and public affairs, but his interest in the geological sciences and the natural resources of the West never lagged, and bore fruit in his later advisory work with the federal Committee on Reconstruction towards the end of World War II.

From 1936 to 1951 Wallace served as Principal and Vice-Chancellor

of Queen's University and his distinguished record in education both here and at Alberta is well known. During the war he was active in the re-establishment of returning veterans, their education and employment in the expanding development of the natural resources of the country.

For many years he was a valued member of the National Research Council and as the first president of the Ontario Research Council he was in large part responsible for the stimulation of research in both the natural and applied sciences, and for a great increase in post-graduate training of science students both in the province and elsewhere by a continuing system of scholarships. He was the first Canadian member of the Board of Trustees of the Carnegie Foundation for the Advancement of Teaching, and represented Canada at the London conference which created UNESCO. As president of the Royal Society of Canada in 1941 he showed a keen insight into the problems facing this and other countries in post-war days and in his address on "Planning for Canada" laid down important principles for the better development, in the democratic way, not only of natural resources but also of the education and culture of the Canadian people.

Among the many and well-merited honors which came to him were the order of Commander of St. Michael and St. George and honorary degrees from twenty universities in Scotland, England, Ireland, Germany, the United States and his adopted land.

After retirement in 1951 Dr. Wallace remained for a time almost as active as before, both in educational affairs of the province, on the Defence Research Board, and as executive director of the Arctic Institute, in which his efforts were an inspiration to all.

What the sciences of mineralogy and geology lost, or might have gained, in the career of this great man, had he chosen otherwise, accrued in full measure to the fields of education, human relations and the public welfare. His mark has been left on many generations of students of three universities, as on all with whom he came in contact—in the field or elsewhere, in this country and abroad. He will be long remembered.

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PRESENTATION OF THE ROEBLING MEDAL OF THE  
MINERALOGICAL SOCIETY OF AMERICA TO  
ALEXANDER NEWTON WINCHELL

PAUL F. KERR, *Columbia University, New York City.*

*Mr. President, Fellows and Members of the Mineralogical Society of America and Guests:*

Some twenty years ago, arrangements were completed by the Council of the Society for the annual award of a medal. The medal was to be awarded for meritorious achievement in the fields of the mineralogical sciences. This medal, now well-known as the Roebling Medal, has been awarded on thirteen occasions. Today we are assembled to honor the fourteenth recipient, Dr. Alexander Newton Winchell.

In honoring Dr. Winchell, the Society is not only recognizing distinguished achievement but is paying homage to American academic tradition in the science of mineralogy. For 37 years the Medalist was Professor of Mineralogy and Petrology at the University of Wisconsin. Before going to Wisconsin he was on the staff of the Montana School of Mines for seven years, and since retiring from the University of Wisconsin eleven years ago he has been a visiting professor both at the University of Virginia and at Columbia University. Even today, while officially enjoying a well-earned retirement, Professor Winchell makes his home at Hamden, Connecticut, close to the academic orbit of Yale University.

Scores of American mineralogists and geologists have been aided in their training through their contact with Professor Winchell in the classrooms of the University of Wisconsin. Much larger numbers in classrooms elsewhere have had the benefit of the excellent textbooks that he has published. It is unlikely that there is a mineralogist either in the United States or Canada who has not been materially aided in his microscopic work by Professor Winchell's textbooks.

In addition to his duties as a professor and as Chairman of the Department of Geology at the University of Wisconsin for six years, the Medalist has found time to write about 100 technical articles. His contributions to the mineralogy of silicate groups are basic. Group analyses of pyroxenes, amphiboles, feldspars, micas, zeolites, chlorites, and scapolite in particular have received his close scrutiny and careful analytical description.

Not content with studies of natural silicates, Professor Winchell has found time to write a book on artificial "minerals" where one will find listed the optical properties of many transparent compounds. He served as chairman of the National Research Committee on accessory minerals

for seven years. It should also be mentioned that his first assignment upon retirement from the University of Wisconsin involved consultation for three years at the American Cyanamid Research Laboratory at Stamford, Connecticut.

The Medalist was the thirteenth president of the Mineralogical Society of America and has always been active in the affairs of the Society, being a regular participant in meetings. His interest in scientific affairs in general has been broad as shown by his membership in at least twelve scientific or professional societies, including a life membership in the American Institute of Mining and Metallurgical Engineers and membership in the American Chemical Society.

We are proud in America to be able to point to certain families that have carried the tradition of natural science from generation to generation. The name Winchell has long been familiar in this respect. It is a matter of interest at least to mineralogists to observe that the family which has so long been identified with the border area of the Great Lakes should now be transplanted to the northern border of Long Island Sound where the traditions of another famous mineralogical family still remain. Professor Winchell's recent appointment as Honorary Fellow in geology at Yale University brings him even closer to the early center of American mineralogy.

A glance at Professor Winchell's remarkable bibliography will show that in 1897 he wrote a six page article on the Koochiching granite. Another glance will show that in 1954 he published a considerably enlarged second edition of the optical properties of organic compounds amounting to 487 pages. Notwithstanding the full years that lie between, and a life span of more than four score years, the Medalist is even more productive today than at the close of the last century when he started.

Mr. President, you honor not only the Medalist, but the Society that makes the award in presenting the Roebling Medal to Alexander Newton Winchell.

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## ACCEPTANCE OF THE ROEBLING MEDAL OF THE MINERALOGICAL SOCIETY OF AMERICA

ALEXANDER N. WINCHELL, 88 Vineyard Road, Hamden, Conn.

*President Hess, Professor Kerr, Fellows and members of the Mineralogical Society of America, and Guests:*

The Mineralogical Society of America is conferring a very great honor in awarding to me the Roebling Medal and it is with sincere and deeply felt appreciation that I take this opportunity to give my thanks to the Council for their very generous estimate of my merits.

Whatever has been accomplished by me is of course due in large part to my training and I hope you will pardon me if I review briefly some parts of it.

Rather indirectly, but still very definitely, I think I was influenced very early in my life by my uncle, Alexander Winchell, who lost his position on the faculty of Vanderbilt University because he accepted the theory of the evolution of man; soon afterward he became professor of geology at the University of Michigan, where he spent the rest of his life. Much more directly was I influenced by my father, Newton H. Winchell, who spent most of his active life as State Geologist of Minnesota, and in that position aided in the development of the vast iron ore deposits of the Mesabi Range. My older brother, Horace V. Winchell, studied at the University of Michigan and later did much toward introducing geology and mineralogy to the mining industry of this country by establishing the Geological Department of the Anaconda Copper Company.

However my first actual study of mineralogy at the University of Minnesota was directed by Charles P. Berkey, whose recent passing away is a source of deep sorrow to us all and a great loss to science. He was called soon afterward to Columbia University and later became known all over the world as the Secretary of the Geological Society of America—an organization which has been most cooperative and helpful to our own society.

I continued the study of mineralogy (especially optical mineralogy) under the guidance of Professor Lacroix at the University of Paris for two years. At that time mineralogists accepted the idea that a mineral was a product of nature having a *definite* chemical composition. This idea was useful as a means of making clear the difference between a mineral and a rock, but it led to the conclusions that (for example) albite was pure sodium aluminum silicate, anorthite was pure calcium aluminum silicate and labradorite was exactly half sodium and half calcium aluminum silicate. But even at that time mineralogists were in doubt about oligo-

clase—was it four parts albite or three parts albite to one part anorthite? Then Tschermak, the famous Vienna mineralogist, proposed the new idea that plagioclase feldspar was a continuous series from pure sodium to pure calcium aluminum silicate. That marked the beginning of a new



ALEXANDER N. WINCHELL

Recipient of the Roebling medal of the Mineralogical Society of America.

era in the history of mineralogy which has led to our present knowledge that nearly all minerals vary in composition.

I have tried to show graphically the relations between variations in composition and variations in optical properties in many minerals. Indeed I have developed more than three-fourths of the hundred and twenty such diagrams which are now in use. In the past such diagrams have been based almost wholly on the chemical analyses and measured

physical properties of minerals found in nature. I have repeatedly warned mineralogists that the diagrams should be considered as approximations rather than highly accurate presentations. This is true even if the chemical analyses and measurements of properties are absolutely correct because natural minerals almost always contain minor variations in composition which are not (and usually cannot be) included in the diagrams, but nevertheless have some effect on the physical properties. As a simple illustration of this condition one may cite plagioclase which almost invariably contains some potassium. However some elements have a much greater effect upon the optical properties than other elements and that has made it possible for our president, Professor Hess, in a recent study of natural clinopyroxenes to obtain results which show remarkably high accuracy as far as the tenor of Fe is concerned. Since minor variations in composition can be eliminated by making artificial minerals it seems clear that they furnish a possible source of greater accuracy. Some excellent work along this line has been done by the staff of the Geophysical Laboratory and others, but much more is needed.

In 1934 the University of Wisconsin granted me a leave of absence which made it possible for me to study x-ray methods briefly with Linus Pauling at the California Institute of Technology, and then go to Manchester, England, to study with W. H. Taylor and W. L. Bragg. Roentgenographic studies are proving very helpful to our science and will undoubtedly continue to do so. Of course you all know that it was Bragg who first taught us how to determine the crystal structure of minerals by x-ray studies, but you may not remember that it was Taylor who first proved that plagioclase is a double (not a single) series.

These are the men, who, during half a century, influenced my studies and work in the field of mineralogy; indeed it is true that without their training my work in mineralogy would have been impossible. Therefore in a certain sense the honor coming to me should really be awarded to them. Please let me repeat how highly I appreciate the honor you have conferred on me by the award of the Roebling medal.



## PRESENTATION OF THE MINERALOGICAL SOCIETY OF AMERICA AWARD TO JULIAN ROYCE GOLDSMITH\*

D. JEROME FISHER, *University of Chicago, Chicago, Illinois.*

*Ladies and Gentlemen of the Mineralogical Society of America:*

It is a matter of statistics that during the school year 1.6 decades back there were 30 students taking my crystallography-mineralogy sequence. Most of these became geologists, but four of them or 13% turned into mineralogists and are fellows or members of this Society, and two of these have received the Mineralogical Society of America award. So far this may look good. But while statistics such as these are exact and correct, their interpretation can be and frequently is controversial. "Figures don't lie, but liars figure." In this particular case it is clear I played no important role in shaping any of these individuals; they took their Ph.D.'s under others. I can only claim that I did not keep them out of mineralogy. As a specific example let us consider today's award-winner. What influence may I have had on him? Well, I try to have my students imbibe a little culture. I emphasize what slaves we are to words and slogans. Who wants to be just a technician; or even just a scientist? I indicate it would be nice not to mis-pronounce the names of common minerals. But today's recipient invariably says peer-ocks-een, not py-rok-seen, though he does say py-rite, not peer-ite. What do you suppose the English natural scientist who devised the reflection goniometer in 1809 W. H. Wollaston (Wool'-ās-tūn) thinks as he looks down on this promising youth who is likely to speak of wō-last'-ton-ite? And like the telephone girls, he says aitch-oh-el, meaning of course aitch-naught-el, though I am sure deep down he realizes that "oh" is the fifteenth letter of our alphabet, not a zero.

The conditions of the Mineralogical Society of America award given this year require that it be based primarily on contributions published after October 1951 and before March 1954 or the 35th birthday of the recipient, whichever came first. However, this work can hardly be isolated from that which preceded it. After his return to the University from Corning Glass where he spent the war years as a research chemist, Goldsmith

\* Goldsmith, Prof. Julian R(oyce), Rosenwald Hall, University of Chicago, Chicago 37, Ill. GEOLOGY. GEOCHEMISTRY. Chicago, Ill., Feb. 26, 18; m. 40; c. 3. S.B., Chicago, 40, Ph.D. (geol.), 47. Asst. petrol., Chicago, 41-42; research chemist, Corning Glass Works, 42-46; asst. petrol., CHICAGO, 46-47, research assoc. GEOCHEM., 47-51, asst. prof., 51-55, ASSOC. PROF., 55-. A.A.; Miner. Soc. Amer.; Miner. Soc. London; Geol. Soc.; Chem. Soc.; Ceramic Soc.; Phase equilibria and crystal chemistry of silicates (feldspars) and carbonates.

carried out his thesis under the supervision of Norman L. Bowen, then Charles L. Hutchinson Distinguished Service Professor of Petrology. This covered a portion of the soda-lime-alumina-silica system containing anor'thite (not an'-orthite), and appeared in 1947. During each of the following two years another paper was published dealing with neighboring portions of this quarternary system.

But by this time Goldsmith had become interested in other than dry silicate-system phase-rule studies, as is shown by his 1950 paper on synthetic Ga-Ge in place of Al-Si feldspars; some of these were prepared in bombs under hydrothermal conditions. Julian succeeded in growing K, Na, and Ca end-member Ga-Ge feldspars, and some partial-substitution Ca-feldspars; on all these he obtained thermal and refractive index data. These crystals were prepared in hopes that single-crystal x-ray study of them would yield conclusions regarding feldspar structure obtainable if at all only with very great difficulty from ordinary feldspars. That this hope was valid was found to be true in later work in collaboration with F. Laves; only this year has appeared the first paper going into details as regards the results of such studies on the Ca feldspars, though an abstract was published in 1951.

Further hydrothermal work with Ca feldspar then resulted in recognition that the hexagonal polymorph which had been identified at the Geophysical Laboratory from material formed under dry conditions around 1250° C. developed in a hydrothermal environment below 375° C. As a by-product of this work, synthetic soda-free thomsonite was produced; also a Ga-bearing form of this material.

During those years of Goldsmith's life when his work resulted in the present award, he had been giving much thought to the order-disorder relations in the feldspars, a problem which Tom F. W. Barth had discussed with him, and the "ease" of crystallization of the various types of silicates. At a time when diffusion as a geologic process was being hotly discussed, he pointed out that the common occurrence of zoned plagioclases had little bearing on this matter in spite of what some had written. The homogenizing of a zoned plagioclase requires a coupled exchange of Na-Ca and Al-Si, and while diffusion of Na-Ca alone might occur without too much difficulty under certain conditions, this was far from the case when interchange of Al and Si was simultaneously involved.

Mr. President, for all this work of great promise, which indeed has already shown fulfillment to a remarkable degree by the publication of at least ten significant papers since his 35th birthday, I take great pleasure in presenting Julian Royce Goldsmith to you as the choice of the committee confirmed by the council to receive the fifth Mineralogical Society of America award.

## ACCEPTANCE OF THE MINERALOGICAL SOCIETY OF AMERICA AWARD BY JULIAN R. GOLDSMITH

JULIAN R. GOLDSMITH, *University of Chicago, Chicago, Illinois.*

*Mr. President, Dr. Fisher, Ladies and Gentlemen:*

When my 12 year old son learned of this award, he asked, what specifically, was to be awarded. When I told him that I was to receive the *American Mineralogist* free, his response was, "Gee whiz, couldn't they give you something like *Life*, or *The Saturday Evening Post*?" If I could not properly transmit my feelings to him, I hope that I can, even without the use of additional words, convey to you the assemblage of emotions associated with being the recipient of this honor. I dare not dwell on other realities such as my unworthiness or the greater worth of the contributions of others, for this would reflect upon the group responsible for the choice. It is perhaps best to change the subject.

It would be even better if I would sit down, but it is difficult to pass up saying a few words to a captive audience. In addition, how else can one get into print in the *American Mineralogist* with a wait of but a few short months? I shall therefore drift into professionalistic "shop-talk."

To fully understand any mineralogical or petrological system, or in general, any homogeneous or heterogeneous system, one must know the equilibrium relations, or states of lowest energy for all temperatures of interest. Single mineral phases, let alone mineralogical assemblages or rocks, can be marvelously complex, and theoreticians are a long way from being able to rigorously compute configurations in systems such as these. Laboratory studies and mineral syntheses have distinct limitations as equilibrium can not always be obtained, and the experimentalist must keep a few natural specimens within sight for a number of reasons. Let us assume that all of our knowledge of the compounds that we call minerals was restricted to that obtained in the laboratory; that we had never seen a rock. The common potassium feldspar of the crust, microcline, would be completely unknown to us, as it has not been synthesized, and we would be quite happy with a sanidine-quartz assemblage as a stable laboratory "rock" that we might call granite.

I have given some thought to the difficulty with which certain substances crystallize relative to others, and have attempted to explain some crystallization phenomena on the basis of what I have called, perhaps unfortunately, the relative "simplicity" of the compounds. In so doing, I have been accused by some of everything from duplicity to "subversity." Permit me to digress to the extent of saying that the word simplicity was not fabricated by me, but can be found in Webster's unabridged,



JULIAN R. GOLDSMITH

Recipient of the Mineralogical Society of America Award.

and merely means simplicity. I prefer to think of it as a series between the end members simplicity and complexity. If a high degree of order, or some other complex configuration coupled with lack of mobility in the system makes it more difficult to produce some phases than others, the "others"

may well appear to be the stable configurations, and fool us into thinking we understand the system under consideration. This might be as real in natural systems as it is in the laboratory, and the lowest energy state in a particular system at or near earth-surface temperatures could be a phase or phase assemblage that no one has ever seen. This difficulty which tends to disappear at higher temperatures obviously cannot be resolved by the back-extrapolation of experimentally determined  $P$ - $T$  curves into frozen regions with the tacit assumption that the equilibrium assemblage is unchanged, or by computation of relative energies of known configurations at lower temperatures. Thermodynamic data, carefully and laboriously obtained at very low temperatures, may show up residual entropies that would be in apparent violation of the Third Law of thermodynamics, and would indicate that other phases are stable, but these data cannot tell us what these phases may be.

One might ask, "So what? Things are as they are and all we are concerned with is knowing under what conditions they got there." This, however, does not really satisfy one's insatiable curiosity. If we are now at the point where I can be considered to be undergoing metaphysical polymorphism, I had surely best sit down, before I bore you to the point where you would not hear me express my deepest thanks for being named recipient of the Mineralogical Society of America Award.



# PROCEEDINGS OF THE THIRTY-SIXTH ANNUAL MEETING OF THE MINERALOGICAL SOCIETY OF AMERICA AT NEW ORLEANS, LOUISIANA

C. S. HURLBUT, JR., *Secretary*

The thirty-sixth annual meeting of the Society held on November 7-9, 1955, at the Roosevelt Hotel New Orleans, Louisiana, was attended by 185 fellows and members. Seventy-three scientific sessions were held at which eighty-two papers were presented.

The annual luncheon of the Society on November 8 was attended by 179 fellows, members and guests. Following the luncheon, the fourteenth presentation of the Roebling Medal was made to A. N. Winchell and the fifth presentation of the Mineralogical Society of America Award was made to Julian H. Goldsmith.

On the afternoon of November 8 the Society was addressed by the Retiring President, Harry H. Hess, on *Some mineralogical and petrological problems of the Stillwater complex*. At the business meeting preceding the presidential address, brief reports were read by the Secretary, Treasurer and Editor.

The 1955 Council of the Society met for seven hours on November 6, the 1956 Council met for three hours on November 9.

For the first time in many years the Mineralogical Society of America did not have its own printed program but the program of the Geological Society of America was sent to all fellows and members. This resulted in a substantial saving to the Society and, from reports so far received, little inconvenience resulted.

The 1954 Council felt that the specifications of the Mineralogical Society of America Award were not stated in a rigorous enough manner and sections 3 and 4 have been revised. The conditions, including the recent changes, under which the award is to be given are as follows:

1. The name of the award shall be the Mineralogical Society of America Award.
2. The award shall be life membership in the Mineralogical Society of America. A certificate as evidence of the award, signed by the President and Secretary of the Society, shall be presented to the recipient at the annual meeting.
3. The award shall be given to an individual for published results of original research constituting an outstanding contribution or series of contributions within the fields of interest of the Society. The candidate's current activity in research shall be taken into account in assessing such contributions.
4. The work for which the award is given must have been published prior to the month in which the candidate's 35th birthday falls. Candidates are limited to persons who shall not have reached the age of 37 before January 1 of the year in which the award is decided upon by the Council.
5. The award shall be made without regard to nationality. Membership in the Mineralogical Society of America is not a necessary prerequisite.
6. The President shall appoint a committee to nominate a recipient. The selection, however, shall be subject to the approval of the Council.
7. The award shall be given annually only if a suitable candidate can be selected by the Council.

COMMITTEES OF THE MINERALOGICAL SOCIETY OF AMERICA FOR 1956

*Nominating Committee for Officers*

B. H. Mason *Chairman*  
E. S. Larsen III  
A. Pabst,  
J. D. Barksdale  
W. D. Keller  
E. N. Cameron

E. W. Heinrich 56-58  
C. L. Christ 56-58

*Program Committee*

H. S. Yoder, Jr., *Chairman*  
(M.S.A. representative on G.S.A. Committee)

*Nominating Committee for Fellows*

T. F. Bates, *Chairman* 56  
J. J. Runner 56  
A. E. Engel 56-57  
R. J. Holmes 56-57  
G. Switzer 56-58  
A. W. Jolliffe 56-58

*Mineralogical Society of America Award Committee*

R. H. Jahns, *Chairman*  
H. T. Evans  
E. E. Wahlstrom  
J. W. Gruner  
O. F. Tuttle  
R. A. Rowland

*Roebling Medal Committee*

G. Tunnell, *Chairman*  
P. F. Kerr  
C. B. Slawson

*Financial Advisory Committee*

E. P. Henderson, *Chairman*  
R. E. Fuller  
(A third member to be appointed by the President)

*Nomenclature Committee*

G. T. Faust, *Chairman* 56  
G. Tunell 56  
C. O. Hutton 56-57  
E. E. Wahlstrom 56-57  
R. S. Cannon, Jr. 56-58  
H. S. Yoder, Jr. 56-58

*Auditing Committee*

J. C. Rabbitt, *Chairman*  
K. J. Murata  
S. Zeffoss

*Board of Associate Editors*

I. Campbell 56  
B. H. Mason 56  
W. F. Bradley 56-57  
F. J. Turner 56-57

*Representatives*

*National Research Council*  
J. D. H. Donnay  
*American Geological Institute*  
A. Montgomery  
E. Ingerson

REPORT OF THE SECRETARY

*To the Council of the Mineralogical Society of America:*

ELECTION OF OFFICERS AND FELLOWS

Five hundred and five ballots were cast in the election of officers, 146 by fellows and 359 by members of the Society. The officers elected to serve in 1956 are:

*President:* Clifford Frondel, Harvard University, Cambridge, Massachusetts.

*Vice-President:* D. Jerome Fisher, University of Chicago, Chicago, Illinois.

*Secretary:* C. S. Hurlbut, Jr., Harvard University, Cambridge, Massachusetts.

*Treasurer:* Earl Ingerson, U. S. Geological Survey, Washington, D. C.

*Editor:* Walter F. Hunt, University of Michigan, Ann Arbor, Michigan.

*Councilors (1956-58):* Chester B. Slawson, University of Michigan, Ann Arbor, Michigan.

Alfred O. Woodford, Pomona College, Claremont, California.

According to the provisions of the Constitution, the following have been elected to fellowship:

Jose Luis Amoros, University of Barcelona, Barcelona, Spain.  
 George William Brindley, Pennsylvania State University, University Park, Pennsylvania.  
 Charles Wesley Chesterman, California State Division of Mines, San Francisco, California.  
 Stephen Edmund Clabaugh, University of Texas, Austin, Texas.  
 James William Earley, Gulf Research & Development Co., Pittsburgh, Pennsylvania.  
 Hugh Richard Gault, Lehigh University, Bethlehem, Pennsylvania.  
 William Thomas Holser, Battelle Memorial Institute, Columbus, Ohio.  
 Howard Francis McMurdie, National Bureau of Standards, Washington, D. C.  
 George Joseph Neuerburg, U. S. Geological Survey, Menlo Park, California.  
 Francisco Pardo, University of Barcelona, Barcelona, Spain.  
 Rustum Roy, Pennsylvania State University, University Park, Pennsylvania.  
 Robert LeGrande Stone, University of Texas, Austin, Texas.  
 John Verhoogen, University of California, Berkeley, California.  
 Alice Mary Dowse Weeks, U. S. Geological Survey, Washington, D. C.

#### MEMBERSHIP STATISTICS

November 1, 1955

	1954	1955	Gain	Loss
Correspondents.....	4	4	0	0
Fellows.....	329	338	14	5
Members.....	931	957	143	117
Subscribers.....	934	952	101	83
	<hr/> 2,198	<hr/> 2,251	<hr/> 258	<hr/> 205

The above figures show a net gain of 9 fellows, 26 members and 18 subscribers. Considering the four groups together, there is a gain of 53, giving a total of 2,251.

During 1955, the Society lost through death three fellows: John P. Marble of Washington, D. C., Charles Palache of Charlottesville, Virginia, and Robert C. Wallace of Ontario, Canada.

Respectfully submitted,  
 C. S. HURLBUT, JR., *Secretary*

#### REPORT OF THE TREASURER FOR 1955

*To the Council of the Mineralogical Society of America:*

Your Treasurer submits herewith his report for the fiscal year beginning August 1, 1954, and ending July 31, 1955.

#### RECEIPTS

Dues and subscriptions.....	\$11,335.25
Sale of back numbers.....	3,203.80
Authors' charges on reprints.....	1,553.43
Interest and dividends from endowment.....	5,086.78
Geological Society of America aid for printing the Journal.....	8,000.00
Advertising.....	780.36
Sale of Index, volumes 21-30.....	22.80
Sale of Index, volumes 1-20.....	15.50
Contributions to Ross-Schaller volume.....	102.00
Premium on check.....	.41
	<hr/>
	\$30,100.33
Cash on hand, August 1, 1954.....	10,544.58
	<hr/>
	\$40,644.91

DISBURSEMENTS

Printing and distribution of the Journal (7 issues).....	\$18,613.75
Printing and distribution of reprints.....	1,350.44
Honorariums to Editor, Secretary, and Treasurer.....	1,437.50
Clerical assistance.....	1,271.49
Postage and express.....	599.82
Printing and stationery.....	338.93
Office supplies and equipment.....	17.17
Program and abstracts (1954).....	695.28
Roebling Medal (1954).....	131.44
New securities purchased.....	7,591.25
Commission on securities.....	74.28
Tax on securities.....	.18
Committee expenses.....	4.27
Expenses of officers to 1954 meetings.....	394.74
Safety deposit box.....	7.15
Refunds.....	16.20
Checks returned.....	17.00
Mineralogical Society of America Award certificate.....	8.00
Reprinting back issues in microtext.....	144.68
Mineralogical Society of America luncheon.....	4.50
Service charges on checks.....	.40
Contributions to American Geological Institute.....	105.00
Telephone and telegrams.....	2.86
Back numbers purchased.....	71.00
	<hr/>
	\$32,897.33
Cash on hand, July 31, 1955.....	7,747.58
	<hr/>
	\$40,644.91

The endowment funds of the Society as of July 31, 1955, consist of the following securities:

BONDS

6M Atlantic Coast Line, 4½.....	\$5,257.50
5M New York Central, 5.....	4,300.00
5M Southern Railway, 5.....	5,743.75
	<hr/>
	\$15,301.25

PREFERRED STOCKS

200 Southern California Edison, 4.88.....	\$5,250.00
100 Union Pacific, 4.....	4,570.25
60 Jones and Laughlin, A, 5.....	4,987.50
55 United States Steel, 7.....	6,946.20
50 Virginia Electric and Power, 5.....	5,942.50
10 Consolidated Edison, 5.....	1,066.64
	<hr/>
	\$28,763.09

## COMMON STOCKS

330 Potomac Electric Power.....	\$4,642.73
200 Greyhound Corporation.....	2,300.00
150 Spancer Kellogg.....	3,775.00
100 Columbus and Southern Ohio Electric.....	2,087.50
100 Diana Stores.....	1,250.00
100 Pittsburgh and West Virginia.....	2,787.50
80 Kroger Company.....	1,990.00
60 United Fruit.....	3,067.50
56 Standard Oil of New Jersey.....	1,444.84
60 American Telephone and Telegraph.....	8,195.32
50 Chesapeake and Ohio.....	2,368.75
50 Phelps Dodge.....	1,975.00
50 Sinclair Oil.....	2,968.75
40 Plymouth Cordage.....	2,050.00
30 U. S. Playing Card.....	2,411.25
24 Public Service Electric and Gas.....	728.40

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 44,042.54

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 \$88,106.88

Respectfully submitted,  
EARL INGERSON, *Treasurer*

## DANA FUND

No disbursements were made during the fiscal year.

Available balance, August 1, 1954.....	\$99.13
Interest recorded, July 25, 1955.....	.12
Available balance, July 31, 1955.....	\$99.25

Respectfully submitted,  
EARL INGERSON, *Treasurer*

## REPORT OF THE AUDITING COMMITTEE

*To the President of the Mineralogical Society of America:*

The Auditing Committee has examined and verified the accounts of the Treasurer of the Mineralogical Society of America for the fiscal year beginning August 1, 1954 and ending July 31, 1955. The securities listed in the Treasurer's report, with all future coupons on the coupon bonds attached, are in the safety deposit box at the 17th and G Street Branch of the Riggs National Bank in Washington, D. C.

Respectfully submitted,  
CHARLES L. CHRIST  
JOHN C. RABBITT  
HOWARD F. McMURDIE, *Chairman*

## REPORT OF THE EDITOR FOR 1955

*To the Council of the Mineralogical Society of America:*

In order that there might be continuity and direct comparison with reports of previous years, the editor's report for 1955 will follow in general the same plan and procedure of



previous years. With five issues published and distributed and the November-December in galley form sufficient data are now available to give a reasonably accurate general survey of the year's activity. When the editor's report appears in print in the March-April issue certain details incomplete and not available at the present time will be included in the final printed report.

In general, it may be stated that the accomplishments represent what might be termed the work of a good normal year with a volume of approximately 1175 pages—about 120 pages larger than the volume for 1954. Two innovations in connection with the 1995 issues might be briefly recalled to mind. The July-August number was shared with our Canadian friends. Part I of that issue contained "Contributions to Canadian Mineralogy" under the able editorship of Professor L. G. Berry of Queen's University. In this section the guest editor assembled 17 interesting papers on various mineralogical subjects and assumed full responsibility for their selection and in seeing the contributions through the press. In Part II of the same issue, 6 other papers were selected from the editor's files. Then, by Council action, the November-December number was dedicated by his colleagues and friends to Professor Emeritus Edward H. Kraus on the occasion of his eightieth birthday (Dec. 1, 1955). This issue contains 18 major and 6 shorter contributions. To those who entertained the view that this action might further delay the printing of manuscripts already on file, I may add that some of the articles from close friends of Dean Kraus that had previously been received were transferred to this number with the hearty approval of the authors concerned.

The current volume, as indicated, will run about 1175 pages—averaging nearly 200 pages for each of the six issues, and contains 85 leading articles. Forty-one additional short papers, appearing under Notes and News, give an overall total of 126 published manuscripts for the year. These contributions were received from 140 contributors associated with 55 different universities, research bureaus and technical laboratories. Forty-six critical book reviews and 11 new minerals described in detail for the first time are a few of the other items of special mineralogical interest.

As in previous years our service during 1955 has extended far beyond our own borders. During the current year 28 manuscripts came from six countries: Australia, Canada, England, Japan, New Zealand and South Africa.

As in the past the Geological Society of America has continued its liberal financial support toward defraying a substantial portion of the printing cost of the Journal. For this assistance the Society is indeed very grateful and expresses its deep appreciation.

And finally a word as to the prospects for 1956. The January-February manuscripts containing 13 main articles and 7 shorter papers are in the hands of the printer at present. In addition at the present time there are on file 16 major and 24 shorter contributions, or a total of 40 papers, aggregating about 425 typed pages, as compared with 66 papers a year ago at this time, aggregating about 900 typed pages, so that substantial progress has been achieved in decreasing the heavy back-log that has caused considerable concern in the past.

It may be recalled that the Council a year ago, in an effort to reduce the period between acceptance and publication, stressed the importance of condensation and rigid critical examination, especially of all major contributions. Your editor in pursuance with this action, in addition to his Board of Associate Editors, called upon 19 readers during the past year. I wish at this time to publicly express my appreciation of this service to the following: L. E. Alexander, G. W. Brindley, R. Denning, J. D. H. Donnay, G. T. Faust, M. Fleischer, C. Frondel, J. W. Gruner, E. W. Heinrich, S. B. Hendricks, H. H. Hess, C. O. Hutton, Earl Ingerson, F. C. Kracek, D. McConnell, A. Poldervaart, L. S. Ramsdell, W. T. Schaller, and R. E. Stevens.

This procedure of very critical inspection has proven very effective in reducing our back-log. To date the critics mentioned and the Board of Associate Editors have examined

56 papers submitted during the year. Twenty-two manuscripts were returned for one reason or another, and 34 accepted, which accounts for the smaller number now on hand compared with the report a year ago at this time.

The accompanying Table 1 summarizes in more detail the distribution of subject matter in Volume 40.

Respectfully submitted,  
WALTER F. HUNT, *Editor*

TABLE 1. DISTRIBUTION OF SUBJECT MATTER IN VOLUME 40

Subjects	Articles	Pages	Per Cent of Total
Leading articles*			
Descriptive mineralogy and paragenesis...	21		
Chemical mineralogy and geochemistry...	21		
Structural crystallography.....	20		
Optical crystallography.....	9		
Geometrical crystallography.....	2		
Petrography.....	3		
Memorials.....	2		
Miscellaneous.....	7		
	85	920	79.6
Short articles.....	41	112	20.4
Notes and news.....	30	7½	
Proceedings of Societies.....	6	58½	
Book reviews.....	46	41	
Abstracts of new mineral names.....	49	17	
Total entries.....	257	1156	100.0
Illustrations.....	249		
Index, Title page, Table of contents.....		27	
Grand total.....		1183	

\* Leading articles average 10.8 printed pages.

#### LIST OF FORMER OFFICERS AND MEETING PLACES

By recommendation of the Council, a complete list of past officers is printed in the proceedings of the annual meeting of the Society:

##### HONORARY PRESIDENTS

Edward S. Dana 1925-1935  
Charles Palache 1949-1954  
Edward H. Kraus 1955-

##### PRESIDENTS

1920 Edward H. Kraus	1923 Edgar T. Wherry
1921 Charles Palache	1924 Henry S. Washington
1922 Thomas L. Walker	1925 Arthur S. Eakle
	1926 Waldemar T. Schaller

1927 Austin F. Rogers  
 1928 Esper S. Larsen  
 1929 Arthur L. Parsons  
 1930 Herbert E. Merwin  
 1931 Alexander H. Phillips  
 1932 Alexander N. Winchell  
 1933 Herbert P. Whitlock  
 1934 John W. Wolff  
 1935 Clarence S. Ross  
 1936 William S. Bayley  
 1937 Norman L. Bowen  
 1938 Ellis Thomson  
 1939 Max N. Short  
 1940 William F. Foshag  
 1941 Frederick E. Wright  
 1942 Arthur F. Buddington  
 1943 John F. Schairer  
 1944 R. C. Emmons  
 1945 Kenneth K. Landes  
 1946 Paul F. Kerr  
 1947 M. J. Buerger  
 1948 M. A. Peacock  
 1949 John W. Gruner  
 1950 George Tunell  
 1951 A. Pabst  
 1952 Michael Fleischer  
 1953 J. D. H. Donnay  
 1954 Sterling B. Hendricks  
 1955 Harry H. Hess

#### SECRETARIES

1920-1922 Herbert P. Whitlock  
 1923-1933 Frank R. Van Horn  
 1933-1934 Albert B. Peck  
 1934-1944 Paul F. Kerr  
 1944- C. S. Hurlbut, Jr.

#### VICE-PRESIDENTS

1920 Thomas L. Walker  
 1921 Waldemar T. Schaller  
 1922 Frederick A. Canfield

1923 George F. Kunz  
 1924 Washington A. Roebbling  
 1925 Herbert P. Whitlock  
 1926 George Vaux, Jr.  
 1927 George L. English  
 1928 Lazard Cahn  
 1929 Edward Wigglesworth  
 1930 John E. Wolff  
 1931 William F. Foshag  
 1932 Joseph L. Gillson  
 1933 Frank B. Guild  
 1934 William A. Tarr  
 1935 Ellis Thomson  
 1936 Harold L. Alling  
 1937 H. V. Ellsworth  
 1938 Kenneth K. Landes  
 1939 Burnham S. Colburn  
 1940 Ian Campbell  
 1941 William J. McCaughey  
 1942 Martin J. Buerger  
 1943 John W. Gruner  
 1944 Harry Berman  
 1945 George Tunell  
 1946 S. B. Hendricks  
 1947 Carl Tolman  
 1948 Adolf Pabst  
 1949 J. D. H. Donnay  
 1950 Ralph E. Grim  
 1951 Michael Fleischer  
 1952 J. D. H. Donnay  
 1953 Sterling B. Hendricks  
 1954 Harry H. Hess  
 1955 Clifford Frondel

#### TREASURERS

1920-1923 Albert B. Peck  
 1924-1929 Alexander H. Phillips  
 1929-1930 Albert B. Peck  
 1930-1940 Waldemar T. Schaller  
 1941- Earl Ingerson

#### EDITORS

1920-1921 Edgar T. Wherry 1922- Walter F. Hunt

#### COUNCILORS

1920 Arthur S. Eakle, Frank R. Van Horn, Fred E. Wright, Alexander H. Phillips.  
 1921 Frank R. Van Horn, Fred E. Wright, Alexander H. Phillips, Austin F. Rogers.  
 1922 Fred E. Wright, Alexander H. Phillips, Austin F. Rogers, Thomas L. Watson.  
 1923 Alexander H. Phillips, Austin F. Rogers, Thomas L. Watson, Esper S. Larsen.  
 1924 Austin F. Rogers, Thomas L. Watson, Esper S. Larsen, Arthur L. Parsons.

- 1925 Thomas L. Watson, Esper S. Larsen, Arthur L. Parsons, William F. Foshag.
- 1926 Esper S. Larsen, Arthur L. Parsons, William F. Foshag, William A. Tarr.
- 1927 Arthur L. Parsons, William F. Foshag, William A. Tarr, Alexander N. Winchell.
- 1928 William F. Foshag, William A. Tarr, Alexander N. Winchell, Ellis Thomson.
- 1929 William A. Tarr, Alexander N. Winchell, Ellis Thomson, Clarence S. Ross.
- 1930 Alexander N. Winchell, Ellis Thomson, Clarence S. Ross, Paul F. Kerr.
- 1931 Ellis Thomson, Clarence S. Ross, Paul F. Kerr, William S. Bayley.
- 1932 Clarence S. Ross, Paul F. Kerr, William S. Bayley, William M. McCaughey.
- 1933 Paul F. Kerr, William S. Bayley, William J. McCaughey, Kenneth K. Landes.
- 1934 William S. Bayley, William J. McCaughey, Kenneth K. Landes, E. P. Henderson.
- 1935 William J. McCaughey, Kenneth K. Landes, E. P. Henderson, J. F. Schairer.
- 1936 Kenneth K. Landes, E. P. Henderson, J. F. Schairer, Arthur F. Buddington.
- 1937 E. P. Henderson, J. F. Schairer, Arthur F. Buddington, Arthur P. Honess.
- 1938 J. F. Schairer, Arthur F. Buddington, Arthur P. Honess, R. C. Emmons.
- 1939 Arthur F. Buddington, Arthur P. Honess, R. C. Emmons, Carl Tolman.
- 1940 Arthur P. Honess, R. C. Emmons, Carl Tolman, D. Jerome Fisher.
- 1941 R. C. Emmons, Carl Tolman, D. Jerome Fisher, Martin A. Peacock.
- 1942 Carl Tolman, D. Jerome Fisher, Martin A. Peacock, Adolf Pabst.
- 1943 D. Jerome Fisher, Martin A. Peacock, Adolf Pabst, C. S. Hurlbut, Jr.
- 1944 Martin A. Peacock, Adolf Pabst, Michael Fleischer, S. J. Shand.
- 1945 Adolf Pabst, Michael Fleischer, S. J. Shand, R. E. Grim.
- 1946 Michael Fleischer, S. J. Shand, R. E. Grim, Joseph Murdoch.
- 1947 S. J. Shand, R. E. Grim, Joseph Murdoch, H. H. Hess.
- 1948 R. E. Grim, Joseph Murdoch, H. H. Hess, Clifford Frondel.
- 1949 Joseph Murdoch, H. H. Hess, Clifford Frondel, Lewis S. Ramsdell.
- 1950 H. H. Hess, Clifford Frondel, Lewis S. Ramsdell, E. F. Osborn.
- 1951 Clifford Frondel, Lewis S. Ramsdell, E. F. Osborn, George T. Faust.
- 1952 Lewis S. Ramsdell, E. F. Osborn, George T. Faust, Victor T. Allen.
- 1953 E. F. Osborn, George T. Faust, Victor T. Allen, C. Osborne Hutton.
- 1954 George T. Faust, Victor T. Allen, C. Osborne Hutton, Felix Chayes.
- 1955 Victor T. Allen, C. Osborne Hutton, Felix Chayes, L. G. Berry.

#### Annual Meeting Places

- |                               |                               |
|-------------------------------|-------------------------------|
| 1920 Chicago, Illinois        | 1938 New York, N. Y.          |
| 1921 Amherst, Massachusetts   | 1939 Minneapolis, Minnesota   |
| 1922 Ann Arbor, Michigan      | 1940 Austin, Texas            |
| 1923 Washington, D. C.        | 1941 Boston, Massachusetts    |
| 1924 Ithaca, New York         | 1942 No meeting held          |
| 1925 New Haven, Connecticut   | 1943 No meeting held          |
| 1926 Madison, Wisconsin       | 1944 No meeting held          |
| 1927 Cleveland, Ohio          | 1945 Pittsburgh, Pennsylvania |
| 1928 New York, N. Y.          | 1946 Chicago, Illinois        |
| 1929 Washington, D. C.        | 1947 Ottawa, Canada           |
| 1930 Toronto, Canada          | 1948 New York, N. Y.          |
| 1931 Tulsa, Oklahoma          | 1949 El Paso, Texas           |
| 1932 Cambridge, Massachusetts | 1950 Washington, D. C.        |
| 1933 Chicago, Illinois        | 1951 Detroit, Michigan        |
| 1934 Rochester, New York      | 1952 Boston, Mass.            |
| 1935 New York, N. Y.          | 1953 Toronto, Canada          |
| 1936 Cincinnati, Ohio         | 1954 Los Angeles, California  |
| 1937 Washington, D. C.        | 1955 New Orleans, Louisiana   |

# RECIPIENTS OF THE ROEBLING MEDAL

Charles Palache, December 1937	William Lawrence Bragg, November 1948
Waldemar T. Schaller, December 1938	Herbert E. Merwin, November 1949
Leonard James Spencer, December 1940	Norman L. Bowen, November 1950
Esper S. Larsen, Jr., December 1941	Fred E. Wright, November 1952
Edward H. Kraus, February 1945	William F. Foshag, November 1953
Clarence S. Ross, December 1946	Cecil Edgar Tilley, November 1954
Paul Niggli, December 1947	Alexander N. Winchell, November 1955

# RECIPIENTS OF THE MINERALOGICAL SOCIETY OF AMERICA AWARD

Orville Frank Tuttle, November 1951	Hatten S. Yoder, Jr., November 1954
Frederick H. Stewart, November 1952	Julian R. Goldsmith, November 1955
L. H. Ahrens, November 1953	

# 1955 ANNUAL MEETINGS AT NEW ORLEANS, LOUISIANA

The meetings of the following Societies were held on Nov. 7, 8, 9, 1955.

- The Geological Society of America—68th
- The Paleontological Society—47th
- The Mineralogical Society of America—36th
- Society of Economic Geologists—36th
- Society of Vertebrate Paleontology—15th
- Association of Geology Teachers

The complete program listed titles and abstracts of 265 papers. Of these 106 seemed to be of special interest to members of The Mineralogical Society of America. Authors and titles of these papers are listed below. Since by Council action the abstracts will not be reprinted in *The American Mineralogist*, references are given to the *Bulletin of the Geological Society of America*, Vol. 66, Number 12, Part 2, December 1955, pp. 1525-1640, where the abstracts of all the papers of the various societies are reprinted, arranged alphabetically by authors.

# PAPERS OF SPECIAL MINERALOGICAL INTEREST

- ISIDORE ADLER AND J. M. AXELROD: *Application of fluorescent x-ray spectroscopy in geochemistry*. Page 1525.
- N. L. ARCHBOLD: *Relationships of calcium carbonate to lithology and vanadium-uranium deposits in the Salt Wash sandstone member of the Morrison formation*. Page 1526.
- WAYNE U. AULT: *Variations of sulfur isotopes in sulfide minerals*. Page 1526.
- ALLAN AUSKERN AND REX W. GRIMSHAW: *Changes to the absorption spectra of kaolinite and monimorillonite during dehydration*. Page 1527.
- PAUL A. BAILLY AND ROBERT R. COMPTON: *Precambrian plutonism at Mineral Ridge, Esmeralda County, Nevada*. Page 1528.
- DONALD R. BAKER: *Stability of magnetite and hematite in a hydrothermal environment from thermodynamic calculations*. Page 1528.
- C. K. BELL: *Some aspects of the geochemistry of gallium*. Page 1529.
- JOSEPH BERMAN AND WILLIAM J. CAMPBELL: *Relationship of composition to thermal stability in the huebnerite-ferberite series*. Page 1530.
- F. DONALD BLOSS: *Farther study of the cleavage tendencies in quartz*. Page 1531.
- F. R. BOYD: *Experimental study of the calciferous amphiboles*. Page 1533.
- LOUIS I. BRIGGS, JR.: *Magmatic differentiation in the Franciscan-Knoxville group of the California Coast Ranges*. Page 1535.
- W. S. BROECKER AND J. L. KULP: *Lamont C<sup>14</sup> Studies*. Page 1535.



- D. BUCKNER AND R. ROY: *Phase equilibria in the system  $\text{CaO-Al}_2\text{O}_3\text{-SiO}_2\text{-H}_2\text{O}$ . II. System  $\text{CaSiO}_3\text{-H}_2\text{O}$  and the influence of  $\text{Sr}^{++}$  in solid solution.* Page 1536.
- WAYNE M. BUNDY: *Petrology of gypsum-anhydrite deposits in southwestern Indiana.* Page 1537.
- F. C. CANNEY, A. T. MYERS, AND F. N. WARD: *Truck-mounted spectrographic laboratory for use in geochemical exploration—a preliminary report.* Page 1538.
- D. R. CARR AND J. L. KULP: *Development in potassium-argon dating.* Page 1540.
- JAIR CARVALHO DA SILVA, JOAN R. CLARK, AND C. L. CHRIST: *Crystal structure of ludwigite,  $\text{Mg}_2\text{Fe}''\text{BO}_3\text{O}_2$ .* Page 1540.
- CARLETON A. CHAPMAN: *Pillow breccia and its significance, Mt. Desert Island, Maine.* Page 1541.
- C. L. CHRIST AND JOAN R. CLARK: *Crystal chemical studies of uranium oxide hydrates.* Page 1542.
- ALVIN J. COHEN: *Color centers in  $\alpha$ -quartz called amethyst.* Page 1543.
- EARL FERGUSON COOK: *Nomenclature and recognition of ignimbrites.* Page 1544.
- GARNISS H. CURTIS: *Importance of Novarupta during eruption of Mt. Katmai in 1912.* Page 1547.
- E. C. DAPPLES AND R. M. MITCHUM: *Petrographic characteristics of sandstones of the Pennsylvanian of the Central Appalachian coal field.* Page 1547.
- MAURICE DEUL: *Mode of occurrence of uranium in the Chattanooga shale.* Page 1549.
- GEORGE W. DEVORE: *Si-Al association in ordered plagioclases.* Page 1549.
- R. C. DEVRIES AND RUSTUM ROY: *Influence of ionic substitution on the stability of micas and chlorites.* Page 1550.
- C. C. DEWITT: *Physico-chemical aspects of mineral deposition.* Page 1550.
- GABRIELLE DONNAY AND J. D. H. DONNAY: *Unusual alkali feldspar intergrowth.* Page 1551.
- GABRIELLE DONNAY AND J. D. H. DONNAY: *Cordylite re-examined.* Page 1551.
- F. DONALD ECKELMANN AND ARIE POLDERVAART: *Geology of the Quad Creek area, Bear-tooth Mountains, Montana-Wyoming.* Page 1552.
- WOLFGANG E. ELSTON: *Volcanic succession and possible mineralization in the Dwyer quadrangle, southwestern New Mexico.* Page 1553.
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#### MEDALS AND AWARDS

*Penrose Medalist*, Maurice Gignoux.

*Arthur L. Day Medalist*, Earl Ingerson.

*Roebbling Medalist*, Alexander N. Winchell.

*Recipient of the Mineralogical Society Award*, Julian R. Goldsmith.

## NOTES AND NEWS

### PRECISION MEASUREMENTS OF THE CELL EDGE OF SYNTHETIC PYRITE

HENRY LEPP, *Univ. of Minnesota, Duluth Branch, Duluth 5, Minnesota.*

In the course of an experimental investigation of the stability ranges of certain iron minerals, a number of synthetic pyrite specimens were produced from chemically pure materials at controlled temperatures. These specimens seemed ideal for an investigation of a possible relation between the cell edge of pyrite and its temperature of formation. Precision measurements were, accordingly, made on three of the specimens representing a range of  $340^{\circ}\text{C.}$  in temperature of formation.

Smith (1942) carried out extensive studies on the physical properties, and more specifically the electrical conductivity of pyrite. He was able to correlate variations in some of these properties with temperature of formation, and it is partly as a result of his work that the present study was undertaken. Peacock and Smith (1941) previously conducted a limited investigation of a possible relation between the temperature of formation and the cell edge of pyrite. They measured two specimens representing the extreme electrical conductivity and hence temperature of formation range, but their measurements failed to show an appreciable variation in the two cell edges.

The present measurements were made on photographs taken with a Picker camera of 10.78 cm. radius using iron radiation. The camera was loaded by the Straumanis method. A reader equipped with magnifier and designed to read directly to 0.005 cm. was used to measure the films. All lines were measured, but only the high  $\theta$  doublets were used for parameter determinations. The final results were plotted against  $\cos^2 \theta$  and extrapolated to  $\cos^2 \theta = 0$  according to the method of Bradley and Jay.

The following specimens were used in this study:

*No. L-55*—prepared by heating at  $540^{\circ}\text{C.}$ , stoichiometric quantities of iron and sulfur powders in a sealed pyrex tube from which the air had been displaced by carbon dioxide.

*No. L-51*—prepared by passing hydrogen sulfide over synthetic siderite at  $350^{\circ}\text{C.}$  Product contained some pyrrhotite.

*No. L-27*—prepared by passing hydrogen sulfide over  $\text{FeCl}_2 \cdot 6\text{H}_2\text{O}$  at  $200^{\circ}\text{C.}$  Product contained some pyrrhotite.

The results of the measurements are compared with published precision cell edge values for natural pyrite in Table 1. For comparison all measurements have been recorded in kX units. Wasserstein (1949) recalculated the Peacock and Smith and the Kerr measurements to these



units, and the writer converted Gordon's measurement from angstrom to kX units.

TABLE 1. COMPARISON OF PYRITE CELL EDGE MEASUREMENTS

<i>Pyrite, Leadville</i>	
Peacock and Smith (analysis $\text{FeS}_{2.0}$ )	$d_0 = 5.4066 \pm 0.0003$ kX units
Kerr, Holmes and Knox	$d_0 = 5.4067 \pm 0.0001$ kX units
Gordon	$d_0 = 5.4070 \pm 0.0003$ kX units
<i>Pyrite, Elba</i>	
Peacock and Smith (analysis $\text{FeS}_{1.98}$ )	$d_0 = 5.4050 \pm 0.0003$ kX units
<i>Pyrite, Synthetic</i>	
L-55 (probably $\text{FeS}_{2.0}$ )	$d_0 = 5.4066 \pm 0.0003$ kX units
L-51	$d_0 = 5.4055 \pm 0.0007$ kX units
L-27	$d_0 = 5.4053 \pm 0.0007$ kX units

The values presented above show that there is a measurable difference in the unit cell dimensions of various pyrite specimens. The close correspondences between the three values for Leadville pyrite and that for specimen L-55 are noteworthy. Peacock and Smith (1941) found by chemical analysis and physical tests that their Leadville specimen had a composition of  $\text{FeS}_{2.0}$ . Since L-55 was formed by heating carefully weighed molar quantities of pure iron and sulfur powders in a sealed tube its composition must have been close to the theoretical  $\text{FeS}_{2.0}$ .

Specimens L-51 and L-27 gave smaller unit parameters than L-55 but their limits of error covered a broader range. The former specimens were prepared by a different method than L-55 and hence one can not conclude from these measurements that the cell dimensions vary with temperature of formation. Peacock and Smith's Elba specimen was found by analysis to be slightly sulfur deficient, and specimens L-51 and L-27 may have been somewhat low in sulfur as indicated by their small pyrrhotite contents. The smaller cell edge values in these specimens may therefore be due to a sulfur deficiency.

*Acknowledgments:* This study constitutes part of a Ph.D. thesis submitted by the writer to the graduate faculty of the University of Minnesota. The interest and aid of Drs. J. W. Gruner and G. Bitsianes are gratefully acknowledged.

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### BORON IN TETRAHEDRA OF BORATES AND BOROSILICATES

WALTER LOEWENSTEIN, *Max Lowenstein & Cia, São Paulo, Brazil.*

Conditions of co-existence and types of condensation of boron and silicon tetrahedra are examined.

It was formerly believed that boron occurred in natural compounds chiefly surrounded by oxygen in triangular co-ordination. Fourfold tetrahedral co-ordination, as determined by Dunbar and Machatschki (1) in danburite, was supposed to be exceptional. More recently, new examples of boron in tetrahedra have been determined, and it seems now that this type of co-ordination is relatively frequent.

Four is an enhanced number of co-ordination of boron towards oxygen, and thus, as was shown by the author in a previous note (2), compounds with boron in tetrahedra can not generally be expected to conform strictly to the Pauling electrostatic valence rule. This had been, in fact, already observed by Berger (3) in the structure of  $B_2O_3$ . While the unfavorable radius ratio makes oxygen bridges between aluminum tetrahedra generally unstable, and hence no such bridges occur in any of the structures of  $Al_2O_3$ , the favorable radius ratio permits boron tetrahedra in  $B_2O_3$  to be linked to each other in such a way that corners are shared by two and even by three tetrahedra.

The maximum number of tetrahedra which can share one corner without sharing edges is four, since the resulting oxygen configuration is cubic close-packed in the ideal case: one central ion is surrounded by twelve others situated at the centers of the edges of a cube. Four cations are tetrahedrally distributed around the strongly polarized central anion. Such a structure occurs in the well-known basic beryllium acetate,  $Be_4O(CH_3CO_2)_6$ . A similar configuration, with four boron tetrahedra sharing one corner, was found by Ito, Norimoto and Sadanaga (4) in boracite. The Pauling electrostatic valence rule is fulfilled if a bond-strength as low as  $\frac{1}{2}$  is attributed to the boron oxygen bond in this particular case. It may reasonably be expected that also the tetraborate ion has a similar structure, and hence in its completely hydrated form has the composition  $[B_4O(OH)_{12}]^{2-}$  or  $[B_4O_7 \cdot 6H_2O]^{2-}$ . Sharing of corners by the oxygen polyhedra which surround the central oxygen ion should lead to the less hydrated forms of sodium borate.

Boron tetrahedra not only can share corners with each other, but also

with silicon tetrahedra, as in danburite. Since the silicon oxygen bond-strength probably can not be reduced to one half in tetrahedral co-ordination, a sharing of one corner by four tetrahedra should under these conditions be impossible. There are, however, two possibilities for condensation of three tetrahedra: two borons and one silicon, and two silicons and one boron. The latter really occurs in the structure of tourmaline, as determined by Donnay and Buerger (5), and by Ito and Sadanaga (6). This is clearly seen by examining the figures in the papers, though it is not mentioned. Figure 1 shows a model of the tourmaline structure. Only the "molecule" with its continuation in the  $z$ -direction is shown;

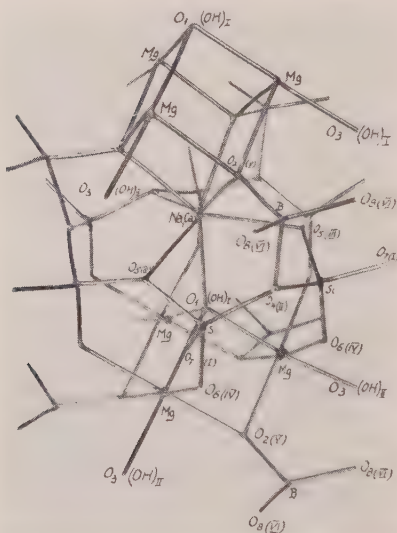


FIG. 1. Model of tourmaline structure.

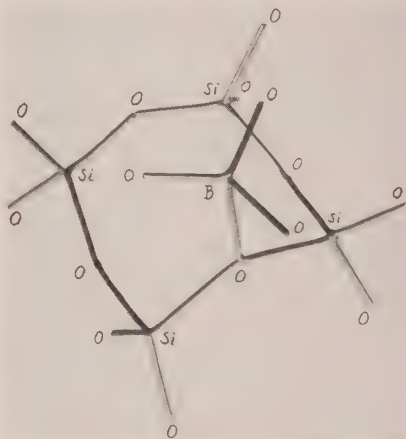


FIG. 2.  $\text{BO}_3 \cdot \text{Si}_4\text{O}_{12}$  group in the structure of axinite.

aluminum atoms are omitted. The boron oxygen  $\text{B}-\text{O}_{4(\text{II})}$  bond may be expected to have a bond-strength of about  $\frac{1}{2}$ , and the distance should accordingly be of the order of 2.2 Å, as in  $\text{B}_2\text{O}_3$  or slightly larger. The bond-strength of the silicon oxygen  $\text{Si}-\text{O}_{4(\text{II})}$  bond should be about  $\frac{3}{4}$ , and the distance, accordingly, slightly larger than the normal 1.62 Å. The boron triangles, really bases of  $\text{BO}_3 \cdot \text{O}$  pyramids, as the distorted tetrahedra are called in the description of the boracite structure by Ito, Norimoto and Sadanaga, should be strongly contracted, and the boron atom should lie very close to the plane of these triangles. Sharing of corners of silicon and boron tetrahedra is the explanation why aluminum can not substitute for silicon in the six-membered silicate ring of tourmaline: an oxygen bridge between aluminum and silicon tetrahedra,

already considerably weaker than the silicon-oxygen-silicon bridge, would break down completely under the weakening influence of the boron oxygen bond. Aluminum tetrahedra thus can not be expected to share corners already shared by boron and silicon tetrahedra.

Another example of the same kind is to be found in the structure of axinite, as determined by Ito and Takéuchi (7). Though the description mentions only separate  $\text{BO}_3$  and  $\text{Si}_4\text{O}_{12}$  groups, these groups are linked to each other by a boron oxygen bond similar to the one existing in tourmaline. The  $\text{BO}_3 \cdot \text{Si}_4\text{O}_{12}$  group is shown in Fig. 2.

It is quite possible that condensation of boron and silicon tetrahedra of the types described is a common feature in the structure of boron silicate glasses, and it may be responsible for some of their properties. It may be expected to occur also in other borosilicates.

*Acknowledgment.* To Professor Martin Julian Buerger, the author wishes to express his gratitude for the suggestion to make a model of tourmaline; and to Messrs. Theodor Dragojevich and Gastão Lorenzini, for their assistance in brazing and photography of the models.

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#### DETERMINATION OF MAGNETITE IN CHRYSOTILE

H. R. SHELL, *Bureau of Mines, Norris, Tenn.*

During work on the beneficiation of chrysotile, the need developed for a method for estimating quantitatively the magnetite content, either originally present or residual. The procedure evolved is given in detail.

A 1-gram sample of chrysotile is cut up by hand or in a Wiley mill to short lengths ( $< \frac{1}{4}$ -inch), placed in a Waring blender containing 500 to 600 cc. water, and disintegrated for 3 minutes. A 0.1 molar solution of aluminum formate is added dropwise until further addition causes no marked improvement in dispersion. 1 cc. is the amount recommended in U. S. Patent 2,661,287 (1). Beating in the Waring blender is continued for 3 minutes to give a total of 6 minutes. This treatment largely disperses the chrysotile into its ultimate fibers, which are only some 200 to 300 Å units in diameter, and dislodges most of the magnetite contained therein. Transfer completely to a 1-liter beaker. (The blender used should be in

good condition and checked on iron-free chrysotile for possible contamination.)

The dislodged magnetite is removed with a strong hand magnet covered with a sheet or bag of polyethylene. The polyethylene should be large enough to act as a rope in letting the magnet into the beaker. The sheet and the magnetite are washed with distilled water, which is allowed to drain back into the beaker. It is estimated that 90 per cent or more of the magnetite present is removed at this time. The magnetite is reserved for combining with a later recovery.

Volume of the solution should now be about 750 to 800 ml. 15 grams of  $H_3BO_3$  are added, and stirring is continued until solution is effected. Then 27 ml. ( $\pm 0.5$  ml.) of 48-per cent HF is added, with vigorous stirring. The beaker is now set in a boiling hot-water bath so that water reaches the level of the liquid inside the beaker. The solution is stirred every 2 minutes, or oftener if desired. After 10 minutes the chrysotile should be largely dissolved. If dispersal has been complete, solution will probably take place in less than 10 minutes. When solution is complete, and in no case over 12 minutes, the beaker should be removed to a cold-water bath, and ice cubes added to cool immediately. As soon as the water is cool enough, immerse the hand magnet as before to collect the magnetite. Remove, wash, and combine the magnetite with the first for determination of iron. HF, HCl, and  $H_2SO_4$ , in platinum, should suffice to dissolve the magnetite for volumetric determination.

Residual flocs of chrysotile generally indicate incomplete dispersion. A second treatment in the steam bath will usually complete the decomposition. Filtering the solution obtained from the steam-bath treatment, to recover magnetite, is not recommended because any non-dispersed material or contaminant will not dissolve, and its iron content would be estimated as magnetite.

The separation depends on three factors:

- (1) The dispersion, or deflocculation, of chrysotile by monovalent anions in combination with  $3^+$  or  $4^+$  cations (1).
- (2) The preferential formation of silicofluoride over fluoborate (2).
- (3) The low reaction rate of magnetite with the complexed fluoride solution.

With 930 milligrams of magnetite (recovered from chrysotile) present, the above procedure resulted in solution of only 16 milligrams, while similarly diluted HF alone, at room temperature, dissolved 82 milligrams of magnetite. Under actual conditions of analysis, solution of magnetite in the boric acid-hydrofluoric acid reagent was less than 0.09 per cent  $Fe_2O_3$  (original sample basis). The amount of magnetite dissolved is a function of amount present, hence with either a small or large magnetite content the results should be accurate.

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## APPLICATION OF DIFFERENTIAL THERMAL ANALYSIS TO IDENTIFICATION OF THE NATURAL HYDROUS FERRIC OXIDES

WILLIAM C. KELLY, *Columbia University, New York City.*

In the course of a recent study of oxidized lead-zinc ore outcrops, the writer attempted to identify the mineral species of hydrous ferric oxide by differential thermal analysis\* of over 150 specimens of gossan "limonites."† These attempts proved unsuccessful, and it was found that thermal curves closely resembling standard analyses (J. L. Kulp and A. F. Trites, 1951) for (1) goethite ( $\alpha\text{-Fe}_2\text{O}_3 \cdot \text{H}_2\text{O}$ ), (2) lepidocrocite ( $\gamma\text{-Fe}_2\text{O}_3 \cdot \text{H}_2\text{O}$ ), and (3) goethite-lepidocrocite mixtures may be produced by goethite alone—the variations depending on the degree of crystallization in the materials tested.

Kulp and Trites observed that *well-crystallized* goethite decomposes directly to  $\alpha\text{-Fe}_2\text{O}_3$  in the temperature range 385–405° C. (heating rate-12°/minute), producing a single strong endothermic peak in the thermal curve of that mineral (Fig. 1A). Lepidocrocite, having a less stable structure, decomposes at lower temperatures. Its curve (Fig. 1B) was described as having an endothermic reaction at about 350° C. (decomposition to  $\gamma\text{-Fe}_2\text{O}_3$ ) followed directly by a variable exothermic peak (phase change of  $\gamma$ - to  $\alpha\text{-Fe}_2\text{O}_3$ ). Thermal analyses of artificial mixtures of the two minerals simply produce composite curves with double endothermic peaks (Fig. 1E); the lower temperature peak representing decomposition of lepidocrocite and the higher temperature peak the decomposition of the more stable goethite.

In analyzing many samples of *poorly-crystallized* goethite, the writer found that the resulting curves (Figs. 1C, 1D) were indistinguishable from standard analyses for lepidocrocite. These goethite samples were of the compact massive and light porous varieties not included by Kulp and Trites in their suite of standard samples. X-ray analyses of 35 of the samples yielded powder patterns with broad and frequently indistinct goethite lines. Under the electron microscope, the materials appeared as moss-like aggregates of goethite with only slight evidences of crystallization even at high magnifications. This type of material constitutes the common yellow or brown "limonite" of widespread and well-known occurrence.

\* The generous advice of Professor Paul F. Kerr regarding use of the thermal analysis equipment at Columbia University was greatly appreciated.

† A general term applying to any natural aggregate of unidentified hydrous ferric oxides lacking *apparent* crystallization.



Mixtures of these poorly-crystallized goethites with some of the original coarsely-crystalline goethite samples tested by Kulp and Trites produced thermal curves (Fig. 1F) matching the standards for goethite-lepidocrocite mixtures. Although these mixture-samples consisted en-

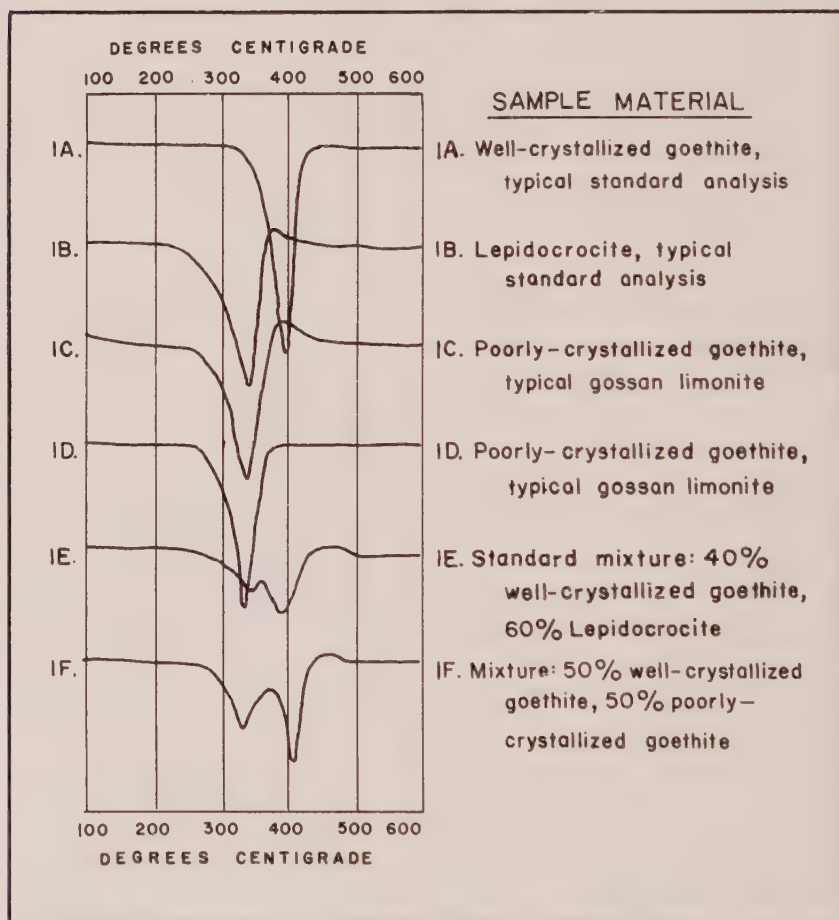


Fig. 1

tirely of goethite and were ground to a uniform particle size (200-mesh), definite differences in the degree of crystallization of the two goethite portions of each sample produced double peaks in the thermal curves. Here, the lower temperature peak represents decomposition of the poorly-crystallized goethite whereas the well-crystallized goethite decomposes at the "standard temperature" around 400° C.

Marked exothermic reactions appeared in the thermal curves of many of the poorly-crystallized goethites (Fig. 1C), but were absent in others (Fig. 1D). X-ray analyses of the heated products chilled at the maximum endothermic decomposition temperatures produced  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> patterns where an exothermic reaction had appeared in the goethite curve and  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> patterns where the exothermic peak was absent. The appearance of a variable exothermic peak due to the phase change of  $\gamma$ - to  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> is usually associated with lepidocrocite, and, as yet, no satisfactory explanation can be offered for its erratic appearance in the thermal curves of these poorly-crystallized goethite samples.

A full report on the present thermal studies is soon to be published by the New Mexico Bureau of Mines and Mineral Resources which aided in the research, but this brief note is submitted to bring out a few of the difficulties encountered with the hydrous ferric oxides. If a curve of the type shown in Figs. 1B, 1C, or 1D is obtained with an unknown, there is uncertainty as to whether the oxide in question is lepidocrocite or poorly-crystallized goethite. A thermal curve with double-peaks (Figs. 1E, 1F) may represent (1) a mixture of goethite and lepidocrocite or (2) a mixture of well-crystallized and poorly-crystallized goethite.

For reliable identification of mineral species of hydrous ferric oxide present in natural aggregates, it is still necessary to resort either to x-ray analysis or to the use of immersion media of high refractive index (H. E. Merwin and E. S. Larsen, 1912).

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#### SYNTHESIS OF ALUMINUM NITRIDE MONOCRYSTALS

J. A. KOHN,\* PERRY G. COTTER, AND R. A. POTTER  
*U. S. Bureau of Mines, Norris, Tennessee.*

#### SYNTHESIS AND GENERAL DESCRIPTION

This note is intended to describe an interesting synthesis of aluminum nitride crystals and some of their properties. The aluminum nitride crystals were obtained incidentally during an attempt to impregnate a sintered carbide compact with alumina. Specifically, a cylindrical WC-TiC-Co compact (5/8 in. diameter  $\times$  5/8 in.) was embedded in

\* Present address: Chemical-Physics Branch, Signal Corps Engineering Laboratory, Ft. Monmouth, New Jersey.

minus-100 mesh, high-purity (99%) alumina (Alcoa, A-14) in a horizontally positioned graphite crucible. The crucible was heated to 2000° C. in a carbon-tube resistance furnace, with a protective atmosphere of helium. Soon after the desired temperature had been reached, the furnace tube, which had been in use for many hours, began to burn out, and the temperature increased sharply to 2100–2200° C. Burning out of the tube allowed an influx of air, which in turn permitted the observed crystallization of aluminum nitride. The latter was formed inside the crucible lid and occurred as pale-blue, prismatic (six-sided) needles,

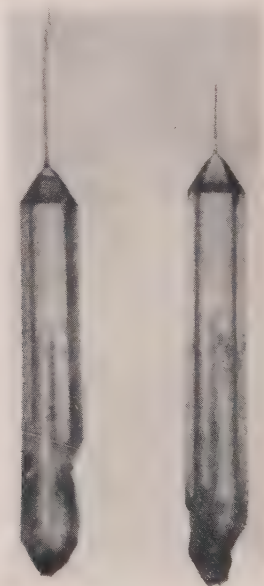


Fig. 1. Aluminum nitride crystals, showing axial "voids" and "antennae" (207 $\times$ ).

approximately 0.03 mm. in diameter and 0.2–0.3 mm. in length. The aluminum nitride crystals, identified as such by x-ray analysis, are shown in Fig. 1. They are clearly doubly terminated, the darker base probably being due to adhering graphitic material. The axial "voids" (negative skeletal structure) and "antennae" (positive skeletal structure) are probably indicative of a rapid rate of growth. An aluminum nitride of this color and habit has been reported by Matignon (1), while Ott (2) described pale-green crystals.

#### PROPERTIES

*X-ray.* Rotation exposures taken about the *c* (length) axis with Cu radiation showed a pattern typical of the wurtzite-type (B4) structure.

Table 1 compares the hexagonal unit cell dimensions obtained with those previously recorded in the literature.

Departure of the axial ratio from the ideal value of 1.633 has recently been confirmed by Jeffrey and Parry (4), who have determined the  $u$  parameter as 0.385 (ideal=0.375). According to the latter authors, this implies that each atom is displaced by 0.05 Å along the  $c$  axis from the center towards the base of its coordination tetrahedron.

*Hardness.*—An attempt was made to obtain Knoop hardness data on the aluminum nitride crystals. Difficulty was experienced in mounting, but a few microindentations were finally placed parallel to the  $c$  axis on the prism face. A  $K_{100}$  value in the low apatite range (Mohs number=5) was indicated. However, these indentations were not considered reliable, owing to the extreme brittleness and small size of the crystals.

Additional data were sought by a microscratch technique. A crystal was mounted upright in balsam, and a lightly weighted slide dragged across the point of the crystal by means of a micromanipulator. The

TABLE 1. AlN UNIT CELL DIMENSIONS

	Ott <sup>2</sup>	Stackelberg and Spiess <sup>3</sup>	Present
$a_0$	3.11 <sub>3</sub> Å	3.104±0.005	3.10 ±0.01
$c_0$	4.98 <sub>1</sub> Å	4.965±0.008	4.96 <sub>5</sub> ±0.010
$c_0/a_0$	1.60 <sub>0</sub> Å	1.600	1.60 <sub>2</sub>

process was observed under a binocular microscope to determine whether or not a scratch was made on the slide. It was found that the crystal was barely able to scratch the glass slide. Since the latter has a Mohs number of 5–5.5, it is probably not in error to quote the hardness of aluminum nitride as in the apatite range. The relative softness of aluminum nitride is in keeping with the fact that no hard material has yet been discovered with a wurtzite-type structure.

*Optical.*—The aluminum nitride crystals were determined to be uniaxial positive, i.e., length slow, since the elongation is parallel to  $c$ . By mounting in thalious chloride, the refractive indices were found to be somewhat lower than 2.247, and a similar comparison with stannic iodide showed them to be above 2.106. A direct birefringence measurement gave an average value of 0.07. These data permitted an estimation of the indices as  $2.13 \pm 0.02$  (omega) and  $2.20 \pm 0.02$  (epsilon). Refinement of these values by comparison with various sulfur-selenium mixtures proved unsuccessful.

*Chemical.*—By means of a small platinum loop affixed to a hot-wire

glass-cutting apparatus, the reaction of aluminum nitride crystals with various hot and cold solutions and with molten salts was observed under the binocular microscope.

The crystals were not affected by the following reagents under both hot and cold conditions: 1-3 HCl, conc. HCl, conc. HNO<sub>3</sub>, 50% HF, and 1-3 NaOH. The crystals did not react with molten NaOH or KOH, although the darker adhering material (probably graphite; see Fig. 1) was rapidly destroyed. The aluminum nitride crystals dissolved *slowly* in molten borax; Al(OH)<sub>3</sub> was precipitated by NH<sub>4</sub>OH from the acidified borax fusion.

The aluminum nitride product described by Matignon (1) was shown by chemical analysis to consist of approximately 1/6 silicon carbide. He stated that the silicon carbide was present either as a concurrent phase or in the form of a solid solution. The crystals studied in the present investigation were too small for chemical analysis. It is felt, however, owing to the relatively low refractive indices (compared to SiC), the relatively low hardness value, and the incompatibility of the structures (no wurtzite form of SiC is known), that there is probably no silicon carbide present.

*Density.*—An approximate density value was known from the literature (1, 2). Accordingly, a suitable heavy-liquid mixture was prepared and sealed in a glass capsule along with several selected aluminum nitride crystals. The capsule was then mounted in a water bath, the temperature of which could be varied. The temperature at which the crystals remained suspended within the capsule was determined, and an experimental density value of 3.24<sub>7</sub> g./cc. at 20° C. was calculated. Ott (2) reported 3.25, while Matignon (1) gave the value at 15° C. as 3.106. The theoretical density calculated from the unit cell dimensions determined herein is 3.30 g./cc. The relatively low experimental value can be accounted for by the negative skeletal structure visible along the needle axes of the crystals (Fig. 1).

*Goniometric.* Several crystals were examined and measurements taken on a two-circle optical goniometer. First order prism faces and first order unit bipyramid faces were observed. Occasionally, reflections could be obtained from a basal pinacoid. Ten measurements of the prism-pyramid interfacial angle ranged from 28°17' to 28°26', averaging 28°21'. The calculated value is 28°24'.

#### ACKNOWLEDGMENT

The authors are grateful to R. A. Hatch, who assisted in the determination of the refractive indices and reviewed the manuscript.



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## MINERALOGY OF THE MIDDLE DEVONIAN TIOGA K-BENTONITE

CHARLES E. WEAVER\*

A thin bed of "meta-bentonite" overlies the Onondaga limestones and cherts of southern New York, central and western Pennsylvania, southeastern Ohio, and West Virginia (Ebright, Fittke, and Ingham—1949), (Flowers—1952), (Fettke—1952).

These bentonite samples<sup>1</sup> have a relatively uniform clay mineral content and are similar in composition to the Ordovician K-bentonites (Weaver—1953). Table 1 contains some 00 $l$  values for the untreated and glycolated less than one micron fractions of the Devonian material and a typical Ordovician sample.

These data indicate that the Devonian K-bentonites consist of randomly interstratified layers of illite and montmorillonite in the approximate ratio of 4:1 to 3:1. X-ray patterns of samples which contain euhedral flakes of "bleached biotite" indicate the presence of chlorite and muscovite. Figure 1 contains x-ray spectrometer patterns of the Albany sample.

The differential thermal curves are also similar to those of the Ordovician K-bentonites (Fig. 1). The molecular water is removed at 100 to 125° C. The hydroxyl water is usually removed in two fractions: 550-600° C. and 675-700° C. The 550-600° C. hydroxyl-endotherm is usually considerably smaller than the 675-700° C. endotherm. The high temperature endotherm occurs at 925° C. and the following exotherm near 1000° C.

Except for more  $Al_2O_3$  and less  $Fe_2O_3$  and  $MgO$ , chemical analysis and mineral formula of the West Virginia sample is similar to that of the

\* Shell Development Company, Exploration and Production Research Laboratory, 3737 Bellaire Boulevard, Houston 25, Texas.

<sup>1</sup> Outcrop samples from Syracuse, New York, were furnished by Dr. N. E. Chute, Syracuse University, and from Albany, New York, by Mr. B. Nelson, University of Illinois. Dr. R. E. Baylor supplied a sample from Chester Co., Pennsylvania, and Mr. W. S. Lytle furnished a sample from Cameron Co., Pennsylvania, and another from Erie Co.

TABLE 1. DEVONIAN K-BENTONITE

	*Calculated 4:1	Ordovician K-Bentonites		Syracuse		Albany		Clinton		Erie		Cameron		West Vir.		Indices	
		1	2	1	2	1	2	1	2	1	2	1	2	1	2	1	2
1	2																
	†11				11.0		11.0				11.0		11.0		11.3		001/001
10.3-10.9	9.5-9.7		10.3-10.9		9.7		10.6		10.3		10.5		10.5		10.6		001/001
5.05-5.1	5.0-5.2		5.0-5.1		5.07		5.03		5.4		5.1		5.07		5.06		001/002
3.31	3.35-3.37		3.30		3.35		3.28		3.30		3.30		3.30		3.30		002/003
							3.24		3.30		3.25		3.35		3.36		003/004
																	003/005

1 = Untreated.

2 = Ethylene glycol.

\* Data from Brown and McEwan (1980).

† Data from Weaver (1955).

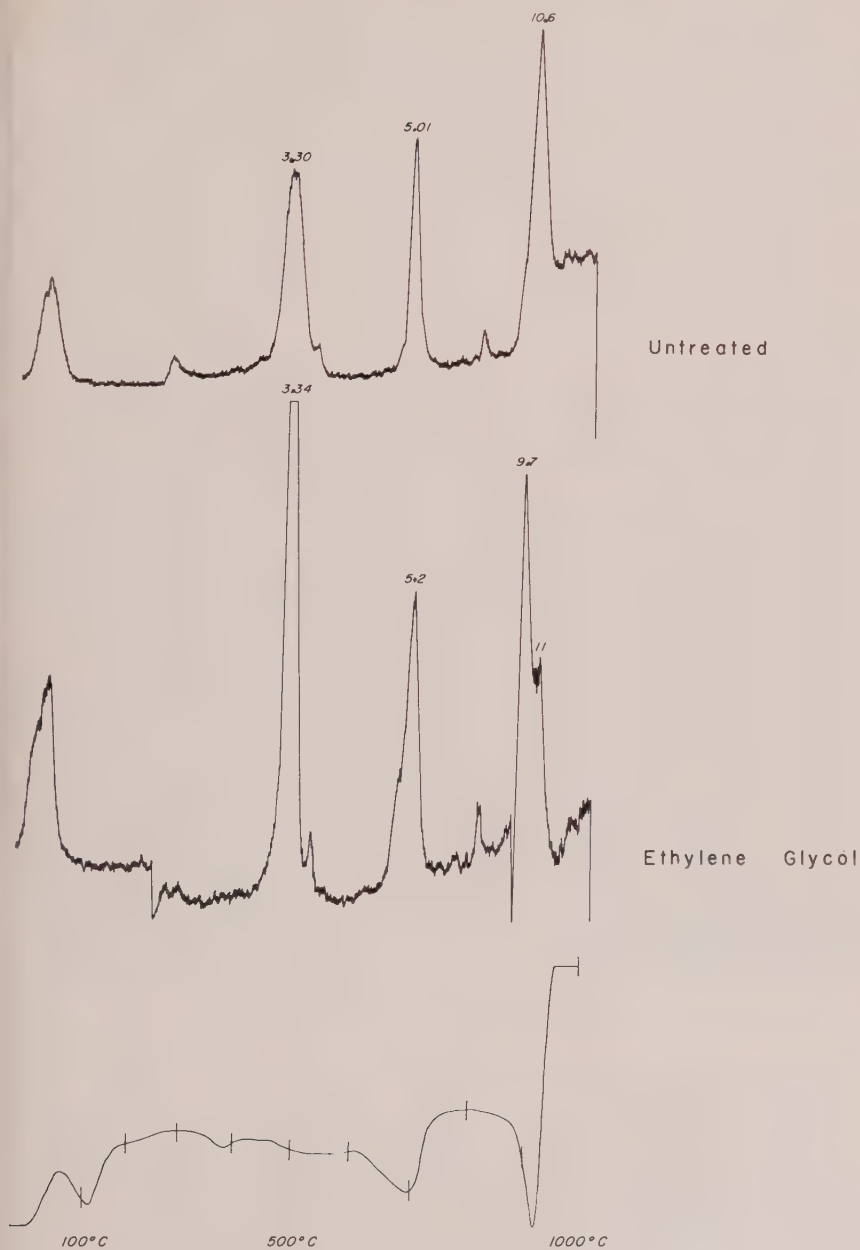


FIG. 1. X-ray spectrometer tracings of untreated and glycolated sample of Devonian K-bentonite from Albany, New York. Differential thermal analysis curve of sample from Syracuse, New York.

Ordovician K-bentonites. The Devonian sample has several per cent more water than is found in the Ordovician samples, but the x-ray and DTA data indicate that this may be an analytical error. As was noted in the Ordovician K-bentonite the potassium cations, presumably between the non expanded layers, and the calcium and sodium cations, between the expandable layers, are present in an approximate ratio of 4:1.

<i>West Virginia</i> <i>Devonian K-Bentonite</i>		<i>Ordovician</i> <i>K-Bentonites*</i>
SiO <sub>2</sub>	47.0	52.7
Al <sub>2</sub> O <sub>3</sub>	28.4	22.1
Fe <sub>2</sub> O <sub>3</sub>	0.15	2.24
MgO	1.92	3.45
CaO	0.91	1.20
Na <sub>2</sub> O	0.4	0.46
K <sub>2</sub> O	5.0	6.06
TiO <sub>2</sub>	0.3	0.4
H <sub>2</sub> O—	11.50	3.78
H <sub>2</sub> O+	4.26	5.79

\* Average of six analyses.



The heavy minerals consist almost entirely of biotite, euhedral zircon and euhedral to subhedral apatite. The suite is similar to that found in the uncontaminated Ordovician K-bentonite beds.

Over an area of at least 40,000 square miles the mixed-layer clays of the Devonian K-bentonites appear to maintain a relatively consistent ratio. This ratio is similar to that of the Ordovician K-bentonites which extend over a much larger area. This 4:1 ratio of non expanded to expanded layers apparently is a relatively stable combination and it may be inferred that the source was an ash with a particular chemical composition (possibly low iron and high potassium and aluminum content) which was altered in a similar environment (carbonate).

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SURFACE ADSORPTION OF  $\text{Cs}^{137}$  IONS ON QUARTZ CRYSTALS\*

STEPHEN ANTKIW, HUGH WAESCHE, AND F. E. SENFTLE,  
*U. S. Geological Survey, Washington 25, D. C.*

A preliminary investigation has been made of the adsorption of radioactive  $\text{Cs}^{137}$  ions on large quartz crystal faces from a solution of  $\text{Cs}^{137}\text{Cl}$ . This was done to determine if there is any relation between imperfections in quartz crystals caused by variations in crystal structure or in growth patterns and adsorption of ions on the crystal surface. If  $\text{Cs}^{137}$  is selectively adsorbed, this technique might be useful in detecting defective

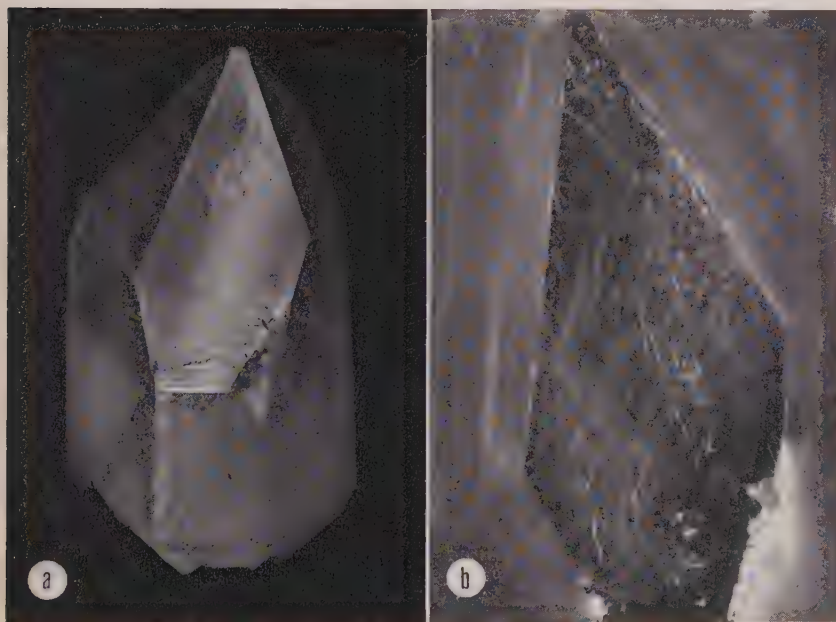


FIG. 1. (a) Natural quartz crystal in white light. (b) Autoradiograph caused by adsorbed  $\text{Cs}^{137}$ .

quartz crystals without destructive testing. Both natural and synthetic crystals were used in the investigation.

Each natural crystal was boiled in nitric acid for approximately 30 minutes to remove surface impurities. Both natural and synthetic crystals were then washed in benzene, alcohol, and de-ionized water. After drying, the crystals were immersed in  $\text{Cs}^{137}\text{Cl}$  solution, the radioactivity of which was about  $5 \mu\text{c}/\text{milliliter}$ , for periods ranging from 20 minutes to 1 hour.

\* Publication authorized by the Director, U. S. Geological Survey.



The synthetic crystals were immersed for the longer periods because adsorption was much less than with the natural crystals regardless of the time of contact with the solution. Autoradiographs made with the two types of commercially available x-ray film gave good results. The films were kept in contact with the natural crystals for 18 hours and with most synthetic crystals for 120 hours. Normal photographic developing techniques were used.

Compared to the synthetic crystals, the natural crystals had many

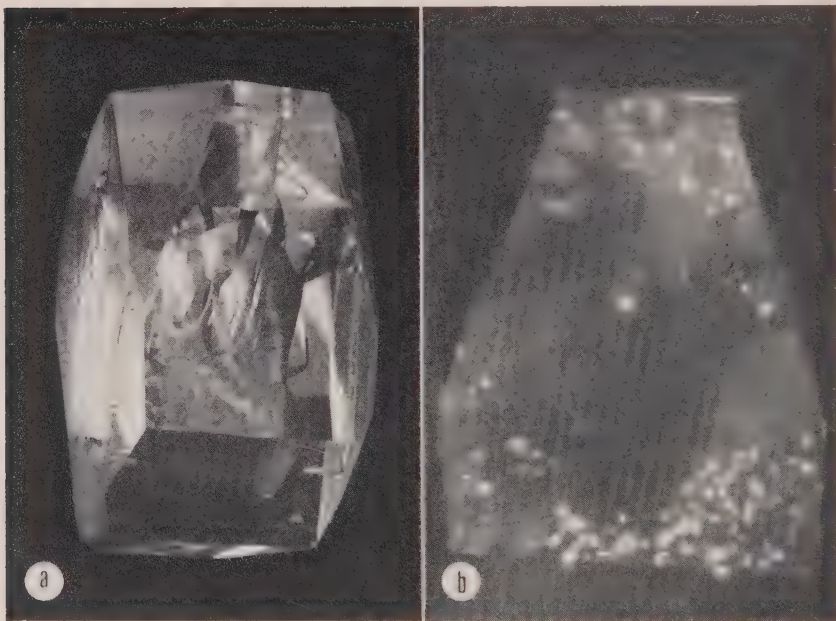


FIG. 2. (a) Synthetic quartz crystal in white light showing spiral-like growth patterns. (b) Autoradiograph showing spots at the apices of the growth patterns and acmite inclusions.

more surface irregularities, such as pits, which adsorbed the  $\text{Cs}^{137}$  ions to a greater extent than the regular crystal surfaces. These irregularities could easily be identified with a hand lens. On the other hand, certain markings were evident that could not be detected with the aid of a lens (see Fig. 1). The major and minor rhombohedral faces showed numerous adsorption lines that seemed to be surface manifestations of growth irregularities complicated by internal fractures and inclusions. These lines may be parallel to regular crystallographic directions, such as  $s$  or  $x$  faces, not exhibited in the outer form of the crystal. It is interesting to note that, although there were visible patches of what appeared to be

electrical twinning and well-developed oscillatory combination lines, no selective adsorption was evident on these crystallographic structures.

As might be expected, the autoradiographs of the synthetic crystals were much clearer than those of the natural crystals. Regular growth lines were common and easily visible to the eye, but produced no effect on the film although surface pits and depressions produced numerous spots (see Fig. 2). Large numbers of surface lineations, which were common in the natural crystals, were not evident on autoradiographs of synthetic quartz. As was evident in natural crystals, electrical twinning did not produce a selective adsorption pattern. In some of the crystals it was evident that what appears to be spiral growth patterns can be emphasized by adsorption. In a number of cases strong adsorption spots on the film coincided with the centers of the spirals (see Fig. 2). This phenomenon seems to confirm the theory advanced by D. Hammond<sup>1</sup> of the Signal Corps Engineering Laboratories that the centers tend to be regular elongated cavities perpendicular to the direction of growth in a growth mechanism of this nature. One of the synthetic crystals also contained artificial inclusions of acmite. The crystal showed a large number of adsorption spots that appeared to coincide with the near-surface acmite inclusions.

This study was undertaken as part of a program conducted by the Geological Survey on behalf of the Division of Research of the Atomic Energy Commission.

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#### ADDITIONAL NOTE ON "BEYRICHITE, A DISCREDITED SPECIES"

In our recent brief note on Beyrichite (*Am. Mineral.*, **40**, 767-770 (1955)) we commented "Unquestionably Schneiderhöhn and Ramdohr are describing violarite." In a letter Professor Ramdohr very properly points out that this statement is obviously in error, violarite being isotropic, and the mineral referred to above being "strongly pleochroic," and further, that the substance in question was probably bismuthinite or emplectite. Both of these have been found by us (*loc. cit.* page 768) and Professor Ramdohr is correct in his identification and we wish to withdraw our erroneous statement concerning it.

CHARLES MILTON, JOSEPH M. AXELROD  
*U. S. Geological Survey,  
Washington 25, D. C.*

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There will be a Symposium on Thermogravimetry and Differential Thermal Analysis at the spring (April) 1956 national meeting of the American Chemical Society in Dallas, Texas. It will be jointly sponsored by the Division of Analytical Chemistry and the Division of Physical and Inorganic Chemistry. The topics to be included relate to the applica-

<sup>1</sup> Oral communication, Frequency Control Review of Technical Progress, April 12-14, 1954. Asbury Park, N. J.

tions of techniques to investigations of reaction mechanisms, kinetic studies, the determination of physico-chemical and thermodynamic properties of inorganic and organic systems, as well as their analysis both qualitative and quantitative. Correspondence should be sent to Dr. Saul Gordon, Pyrotechnics Chemical Research Laboratory, Picatinny Arsenal, Dover, New Jersey.

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The American Iron and Steel Institute Regional Technical Meeting Award for 1954 was presented to Dean E. F. Osborn of The Pennsylvania State University, at the annual general meeting of the Institute in New York City on May 25. The medal was awarded to Dr. Osborn for his paper "Phase equilibrium studies in steel plant refractories systems" which was presented during the October Regional Technical Meeting held in Pittsburgh.

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Dr. G. W. Brindley of The Pennsylvania State University has been named professor of solid state technology and head of the newly named Department of Ceramic Technology, effective July 1. He has been research professor of mineral sciences since his appointment to the faculty of the college on July 1, 1953. Dr. Brindley has also been appointed associate editor of the *A.S.T.M. Index of X-Ray Diffraction Patterns for the Analysis of Materials*.

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A School for Advanced Study is to be established at the Massachusetts Institute of Technology. An opportunity will be afforded for advanced study beyond the level of the graduate school and the doctor's degree. Professor Martin Buerger has accepted the appointment as Director of the School for Advanced Study and also the appointment as Institute Professor.

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Word has been received of the death of Shukusuke Kôzu, professor emeritus of mineralogy and petrology of Tohoku University, Tokyo, Japan, on Feb. 11, 1955, at the age of 74 years.

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## CORRECTIONS

The paper on alluaudite (Nov.-Dec. 1955 issue) should be corrected as follows:

Page 1102—in line no. 16, the figures in columns 5 and 6 should be interchanged; and in line no. 23, the figure in column 4 should be 2.0967 (not 2.1361).

Page 1103—in line no. 24*b*, the figure in column 4 should be 1.9612 (not 2.0481); in line no. 26, the index on the right should be (312) [instead of (3I2)]; and in line no. 28, the spacing on the right should be 1.817 (not 1.813).

Page 1104—The dashed line on the right side of Fig. 2 is the trace of (100).

Page 1108—line 10 from base: instead of "850/860°," read "higher."

## BOOK REVIEWS

PETROGRAPHIC MINERALOGY, by ERNEST E. WAHLSTROM. Pages vi+408, 1955. John Wiley & Sons, Inc., New York. Price \$7.75.

The purpose of this attractive booklet is according to its author to bridge the gap between the books that deal with the determinative procedures of petrography and those dealing with the description and classification of rocks. There is therefore given in this one volume a comprehensive review of techniques, descriptions of minerals, determinative tables, and summaries of classifications of rocks—igneous, sedimentary, and metamorphic. It gives universal stage and petrofabric techniques. Despite the inclusion of this advanced matter, the book has been designed for a one semester course for beginners and for geologists and mineralogists who desire to brush up on what they have forgotten about petrography and petrology.

Chapter 6 of 111 pages gives the properties, both megascopic and microscopic, of the rock-making silicates. It is not clear why the "Physical properties in hand specimens" is sandwiched in between "Crossed polarizing prisms" and "Interference figure," as on p. 186 and elsewhere. The minerals described include such difficultly identifiable minerals as merwinite, mullite, humite, and others, but the descriptions are far too condensed to be helpful. Beginners would appreciate more information on the points of dissimilarity between minerals that resemble one another more or less. It is a cause for astonishment to me, even after thirty years' teaching experience, how many minerals "look alike" to beginning students in microscopical petrography and even to students not so near beginning.

It may be suggested that the "Tables for Mineral Identification," comprising Chapter 8, pp. 235–283, could helpfully be improved by adding a column giving the sign of elongation of the principal section of the minerals. It is a valuable diagnostic property which is easily learned by beginners.

Some of the slips that apparently are inevitable in all first editions appear at various places. Pyroclastic rocks are considered to be igneous rocks on p. 304 but on pp. 355–357 they are described as sedimentary rocks. Although the author believes that the best scheme to follow in megascopically classifying rocks is that proposed by Cross, Iddings, Pirsson, and Washington in 1903, nevertheless he greatly changes the meaning of one of their principal terms "phanerite" by redefining it as a rock "sufficiently coarse-grained to permit at least half of the mineral components to be seen with the unaided eye."

Under "lamprophyres" are brought up from the vasty deeps of petrology into the twilight a large number of highly specialized rock names. Kamperite from Broegger (1921) is one, but the essence of Broegger's definition is not given. No beginning student nor geologists or mineralogists "who occasionally need instruction or refreshment in systematic petrography" for whom the book is confessedly written could be trusted to apply any of these terms correctly without adequate development of the underlying principles. The classification of the lamprophyres is reminiscent of the early editions of Rosenbusch, the founder of the lamprophyre concept, but in his later editions Rosenbusch greatly improved on the earlier versions.

Many fine and effective illustrations adorn the book. Another notable feature is that each chapter closes with a valuable list of selected references, wholly up-to-date, and which should be of much service to those instructors who shall attempt to put across in one semester the large amount of material compressed into this volume.

ADOLPH KNOFF,  
*Stanford University, Stanford, California*



SOUTHERN AFRICA, A GEOGRAPHICAL STUDY: Volume I, Physical Geography, by JOHN H. WELLINGTON. New York: Cambridge University Press, 1955. xxiv+528 pages, 50 plates, 4 colored maps in pocket. Price, \$12.50.

Volume II of this work appeared before Volume I and in its review (*Am. Mineral.*, 40 p. 1150, 1955) the areal coverage and scope are indicated. Volume I is divided into three parts: (1) Structure and Surface, 190 pages; (2) Climate, Vegetation and Soils, 148 pages; and (3) Hydrography, 170 pages. Much of the information is generalized on four large colored maps. These are on a scale of approximately one to four and a half million and show (1) Geological Outlines, (2) Altitudinal Zones, (3) Mean Annual Rainfall, and (4) Vegetation.

The geology of the region is treated very sketchily in Chapter 1 of Part I (24 pages). Three chapters (91 pages) are devoted to the physiographic regions; one chapter (28 pages), to the seas; and one chapter (48 pages), to the coasts. This is perhaps a logical proportion for a geographical study, but gives only the barest outline of the geology. However, there are 44 references to more detailed and comprehensive articles on various aspects of the geology so that the interested reader who has access to a good library can fill in needed details.

The two volumes constitute a delineation of the more important geographical aspects of the subcontinent as described in publications up to the mid-century, together with many hitherto unpublished facts gathered by the author in 34 years of field work.

The work is well bound, very attractively illustrated and withal is an important contribution to the literature on Southern Africa.

EARL INGERSON,  
*U. S. Geological Survey, Washington, D. C.*

QUANTITATIVE SPECTROCHEMICAL ANALYSIS OF SILICATES, by L. H. AHRENS. Addison-Wesley, Cambridge, Mass., 1955, 122 pages, 30 figures, 7 plates. Price, \$3.75.

This monograph is primarily a compilation and unified discussion of a number of analytical methods published by Ahrens and his coworkers in several journals during the past five years. A scheme for determining the major and minor elements of silicate rocks and minerals is presented in nine chapters entitled: Introduction, Miscellany, Standards, Log normal distribution of spectrochemical error, Alkali metals, Volatile elements, In-volatile elements, Fluorine, and Common elements. No attempt is made to cover the literature except that dealing with the determination of the common elements.

Recommended values for the percentage composition of two rock samples, granite G-1 and diabase W-1, are given in chapter 3, and a method of using these samples as spectrographic standards is described. For the minor elements of these samples, the term "tentative values" rather than "recommended values" would have more strongly reminded the reader that the values are based on limited data and that much more research is needed before these samples can be considered in the same category as the standard materials issued by the National Bureau of Standards.

Chapter 5, dealing with the determination of alkali metals, is by far the longest (27 pp.) and contains much useful data. The treatment of the other elements is less detailed.

The book conveniently summarizes the experience of Ahrens and his colleagues during a fruitful five-year period at the Cabot Spectrographic Laboratory of M.I.T. and is a valuable addition to the specialized literature of the field.

K. J. MURATA,  
*U. S. Geological Survey, Washington 25, D. C.*



RÖNTGENSTRUKTURANALYSE VON KRISTALLEN, EINE ELEMENTARE EINFÜHRUNG, by RUDOLF KOHLHAAS AND HELMUT OTTO. Berlin, Akademie-Verlag, 1955, ii+212 pp., 163 figs., 21 tables, 17×24 cm. Price 23 DM.

According to the authors of this book, there exists today in Germany no traditional school of crystal structure analysis despite much good work done there in this field. They feel that this is due partly to the lack of introductory textbooks (in German) suitable for the use of students of chemistry, mineralogy, geology, and petrography. It was to help fill this gap that the present book was written. Because of the type of reader for whom the book is intended, the scope of the material covered is restricted, and in particular, the use of specialized mathematics avoided. A good idea of the contents of the book is gained from the chapter headings which are (in English translation):

- I. Fundamentals of Crystallography (62 pp.)
- II. Production and Properties of X-Rays (12 pp.)
- III. The Determination of the Elementary Cell (16 pp.)
- IV. Indexing, Aids and Procedures (47 pp.)
- V. Determination of the Symmetry of the Elementary Cell by X-Rays (5 pp.)
- VI. The Content of the Elementary Cell (60 pp.)
- VII. The Powder Method of Debye-Scherrer-Hull (7 pp.)

The plan of the book, and the level of presentation are somewhat like those used in C. W. Bunn's "Chemical Crystallography" (Oxford University Press, 1946), although the present volume is much less comprehensive. In general, statements of principles are made clearly and simply but without derivation from more elementary ideas. For example, the fact that crystals can have only (1-), 2-, 3-, 4- and 6-fold rotation axes of symmetry is stated without the proof that this follows from the existence of the lattice. This method of presentation is, of course, entirely appropriate to the present volume.

Fourier methods of crystal structure analysis and analytical applications of the x-ray powder method are not treated. However, a list of reference books covering these and other topics is given.

There are some aspects of the presentation with which one might differ. For example, one finds again the use of the idea of the "interpenetrating lattice," a concept apparently dear to the hearts of German crystallographers. This is an obsolete and unnecessary device and should be discarded. The Schiebold-Sauter method of photographing the reciprocal lattice is treated at some length, whereas the related but much more satisfactory Buerger precession method is given only two short paragraphs; moreover, the latter method is wrongly grouped with the Laue method. There are a few minor errors scattered throughout the book.

Despite the criticisms made above, it is clear that the authors have done an excellent job in fulfilling their stated purpose. The treatment, in general, is lucid and sufficiently comprehensive. The book contains a large number of very good illustrations of the type so necessary to this kind of subject. Numerical examples of problems are given where appropriate, and there are many tables and charts. As is usual with them, the publishers have done an excellent piece of work on the physical makeup of the book.

The book can be recommended for those students seeking a good, elementary approach to the procedures of crystal structure analysis, written in German. For English-speaking students the excellent book of Bunn (mentioned above) is available.

C. L. CHRIST,

*U. S. Geological Survey, Washington 25, D. C.*

## NEW MINERAL NAMES

### Minguzzite

CARLO L. GARAVELLI, Un nuovo minerale tra i prodotti secondari del giacimento di Capo Calamita (Isola d'Elba). *Atti accad. nazl. Lincei, Rend. classe sci fis. mat. e nat.*, **18**, 392-402 (1955).

Small green to yellow-green tabular crystals were found associated with humboldtine,  $\text{FeC}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$ , in limonite at Cape Calamita, Elba. Analysis (mean of 5 partial) gave:  $\text{Fe}_2\text{O}_3$  16.05,  $\text{K}_2\text{O}$  26.00,  $\text{C}_2\text{O}_3$  42.13,  $\text{H}_2\text{O}^+$  11.35,  $\text{H}_2\text{O}^-$  1.60,  $\text{Al}_2\text{O}_3$  0.10,  $\text{FeO}$  2.28,  $\text{MgO}$  0.10,  $\text{CaO}$  0.03, insol. in  $\text{HCl}$  0.10; sum 99.74%, which corresponds to  $\text{K}_3\text{Fe}'''(\text{C}_2\text{O}_4)_3 \cdot 3\text{H}_2\text{O}$  with 6% humboldtine, 1.7% goethite, and 1.6% excess  $\text{H}_2\text{O}$ . The mineral is readily soluble in water. The crystals are tenths to hundredths of a millimeter in size. The forms  $b \{010\}$ ,  $e \{11\bar{1}\}$ ,  $\sigma \{111\}$ , and  $m \{110\}$  were found. Goniometric measurements agreed well with those of Knaggs, *J. Chem. Soc. London*, **121**, 2069 (1922), on artificial crystals, which Knaggs reported to be monoclinic,  $a:b:c = 0.9916:1:0.3895$ ,  $\beta = 94^\circ 13\frac{1}{2}'$ . Cleavage perfect (010). Luster vitreous.  $G = 2.080$  (mineral), 2.092 (synthetic). Optically biaxial, negative, with  $n_s \alpha = 1.498$ ,  $\beta = 1.554$ ,  $\gamma = 1.594$ ,  $2V = 78^\circ$  (calcd.), pleochroic with X yellow-green, Z intense emerald-green. Crystal grown from solution had  $\alpha = 1.501$ ,  $\beta = 1.555$ ,  $\gamma = 1.597$ . X-ray powder data are given; they agree well with those in the *A.S.T.M.* file for potassium ferric oxalate trihydrate.

The name is for the late Carlo Minguzzi, professor of mineralogy at the University of Pavia.

MICHAEL FLEISCHER

### Limaite

J. M. COTELO NEIVA, Pegmatitos com cassiterite e tantalite—columbite de Cabraço (Ponte do Lima—Serra de Arga). *Pubs. Museu e Lab. Mineral. e Geol. e Centro Estudos Geol., Univ. Coimbra, Mem. e Noticias*, No. **36**, 59 pp. (1954)

J. M. COTELO NEIVA, A. RIMSKY, AND A. SANDRÉA, Sur une variété de gahnite stannifère de Cabanas (Portugal). *Bull. soc. franc. minéral. crist.*, **78**, 97-105 (1955).

Pegmatitic veins cutting mica schists contain quartz, albite, muscovite, cassiterite, topaz, and yellow-brown octahedra resembling pyrochlore, 0.5 to 2.7 mm.  $G = 4.47$ ,  $n = 1.81 \pm .003$ , birefringence 0.004-0.005. Analysis on 100 mg. gave  $\text{Al}_2\text{O}_3$  45,  $\text{ZnO}$  32,  $\text{SnO}_2$  13.5,  $\text{Fe}_2\text{O}_3$  3,  $\text{MgO}$  2,  $\text{MnO}$  0.1,  $\text{CaO}$  0.47,  $\text{SiO}_2$  3; sum 99.07%. Spectrographic analysis showed traces of Nb, Be, Sc, Ga, and Ge, no Ni, Cu, Cr, Co, Ti, P, and B. The x-ray powder data are similar to those of spinels, but indicate a superstructure,  $a$  32.20 kX. This unit cell contains  $(\text{Al}_{113}\text{Fe}_6\text{Si}_6)(\text{Zn}_{50}\text{Sn}_{12}\text{Mg}_6)\text{O}_{256}$ . Calcd. density 4.85 (measured 4.47). The mineral is considered to be intermediate between gahnite and nigerite.

The name is for the locality, which is near Ponte do Lima, Portugal. The name is not used in the second paper.

M. F.

### Stillwellite

J. McANDREW AND T. R. SCOTT, Stillwellite, a new rare-earth mineral from Queensland. *Nature* **176**, No. 4480, 509-510 (1955).

Stillwellite occurs in quantity in crystals up to 5 mm. in diameter as a major constituent of radioactive ore from the Mary Kathleen Lease, 34 miles east of Mt. Isa, northwest Queensland, Australia. It is associated with much allanite and with garnet as a metasomatic replacement of metamorphosed calcareous sediments. Uraninite is dispersed in allanite and stillwellite.

Analysis of a concentrate containing 96–98% stillwellite gave as major constituents  $R_2O_3$  58.4 (R=lanthanons),  $B_2O_3$  11.5,  $SiO_2$  20.1, CaO 3.96,  $P_2O_5$  2.58%. (This gives a sum of 96.54%, but the analysis is said to total 100.3%. M.F.)  $Ce_2O_3$  constitutes 51% of the total lanthanon oxide, which contains not more than 5% of heavy lanthanons. Allowing for the presence of a small amount of a phosphate mineral, this leads to the formula  $(R, Ca)BSiO_6$ .

Stillwellite is colorless in thin section. Optically uniaxial positive to biaxial positive with  $2V$  up to  $6^\circ$ . Zoning was noted in thin section;  $\omega$  (Na)  $1.775 \pm .003$  to  $1.784 \pm .003$ , birefringence 0.018–0.020.  $G.=4.57$ . X-ray study showed stillwellite to be hexagonal rhombohedral, space group  $C 3 m$ ,  $C 32$ , or  $C\bar{3}m$ ; the unit cell has  $a=6.85$ ,  $c=6.64$  Å,  $Z=3$ .

The name is for Frank Leslie Stillwell, Australian mineralogist.

### Väyrynenite

A. VOLBORTH, Phosphatminerale aus dem Lithiumpegmatit von Viitaniemi, Eräjärvi, Zentral-Finnland. *Ann. Acad. sci. Fennicae*, Ser. A, III Geol.-Geogr. No. 39, 90 pp. (1954).

A preliminary announcement was abstracted in this journal, 39, 848 (1954). The following additional data are given. The mineral occurs with herderite, hurlbutite, beryllonite, microcline, and muscovite (gilbertite), also in morinite. Color rose-red, transparent, luster vitreous.  $H.=5$ . Cleavage (001). Analysis on 275 mg. gave FeO 4.59, MnO 30.57, CaO 1.82,  $Li_2O$  trace,  $Na_2O$  1.42,  $K_2O$  1.18, BeO 12.10,  $Al_2O_3$  2.45,  $P_2O_5$  40.36,  $H_2O^-$  0.08, insol. 0.78,  $H_2O^+$  (by difference) 5.00, F present, sum 100.35%, corresponding to  $BeMnPO_4(OH, F)$ . X-ray powder data are given; the strongest lines are for two samples in  $kX$  3.41, 3.44 10; 2.83, 2.86 10; 2.62, 2.66 9, 8; 4.39, 4.43 5, 6.

M. F.

### Gearksite

I. F. GRIGOR'EV AND I. DOLOMANOVA, Gearksite, a new water-containing calcium-aluminum fluoride mineral. *Trudy Mineralog. Muzeya Ada. Nauk. S.S.S.R.*, No. 3, 93–96 (1951); from an abstract by Wilhelm Eitel in *Chem. Abstracts*, 49, 13840 (1955). Chemical analysis gave:  $Al_2O_3$  44.42, CaO 16.36,  $H_2O^+$  15.39, F 37.86% (this gives a sum of 114.03 minus 15.94 (O=F<sub>2</sub>)=98.09%. M. F.), corresponding to  $CaAl_3(F, OH)_{11} \cdot H_2O$ . Spectrographic analysis showed weak lines of Fe, Sr, As, Mn, and Ni and traces of Pb, Zn, Na, Co, and Ti.

The mineral occurs in dense white kaolinite-like aggregates,  $n$  about 1.458, birefringence less than 0.009. Differential thermal analysis showed strong endothermal peaks at  $350^\circ$ ,  $490^\circ$ , and  $850^\circ$  and a slight exothermic peak at  $730^\circ$ . The x-ray powder pattern differs from those of gearksutite and creedite.

The mineral occurs in the oxidation zone of deposits in Transbaikal where slates and sandstones have been altered by F-rich hydrothermal solutions. Gearksite is said to be a supergene replacement product of topaz and fluorite. Other associated minerals are zinnwaldite, amazonite and secondary halloysite, montmorillonite, and sellaite.

M. F.

### Betekhtinite

ARNO SCHÜLLER AND ERIKA WOHLMANN, Betehtinit, ein neues Blei-Kupfer-Sulfid aus den Mansfelder Rücken, *Geologie*, 4, No. 6, 535–555 (1955).

The mineral occurs in veins cutting the Mansfeld copper shale. It occurs in irregular masses cut by bornite, chalcopyrite, and galena, and as pure needles associated with calcite. mostly a few tenths of a mm. in diameter, but reaching 2 cm. in length and 5 mm. in

breadth. Some needles have overgrowths of native silver. Analysis by Wohlmann gave: S 17.25, Pb 19.20, Cu 61.39, Fe 1.83, Bi, As, Sb absent; sum 99.94%. Spectrographic traces of Ag and Te were found. This corresponds closely to  $(\text{Cu}, \text{Fe})_{11}\text{PbS}_6$ , but the authors prefer the formula  $\text{Cu}_{10}(\text{Fe}, \text{Pb})\text{S}_6$ .

From  $x$ -ray study, the lattice constants are  $a=3.85$ ,  $b=14.67$ ,  $c=22.8$  Å. Orthorhombic, symmetry class  $C_{2v}$  or  $D_2$ . (This gives a unit cell content containing 4.47 Pb, 46.5 Cu, 1.6 Fe, and 25.9 S. M. F.)  $X$ -ray powder data are given for 5 samples, with somewhat variable results; the one chosen by the authors had  $d$  spacings 2.955 and 1.835 strong, 1.953 and 1.764 medium strong, 3.10 and 2.368 medium. These differ from data for wittichenite, emplectite, and enargite. (No comment is made on the fact that the authors' data for these minerals differ markedly from data in the literature. M. F.).

The mineral polishes well; it is a little harder than bornite. It is a bright cream color parallel to  $c$ , stronger yellowish-cream perpendicular to  $c$ ; it becomes duller on exposure. Reflectivity in air high. Strongly anisotropic.  $G=6.14$ .

The name is for A. G. Betekhtin, Russian mineralogist and economic geologist.

DISCUSSION: The name alisonite (1859) was given to material somewhat similar in composition, presumed to be a mixture. See *Dana's System*, 6th Ed., p. 51.

Betekhtinite appears to be a valid mineral, but further study would be desirable.

M. F.

### Ericaite

F. HEIDE, Über bemerkenswerte Borazitvorkommen in den Kalilagern des Südharzbezirkes. *Chemie der Erde*, **17**, 211–216 (1955).

ROBERT KÜHN AND INGEBURG SCHAAKE, Vorkommen und analyse der Boracit- und Ericaitekrystalle aus dem Salzhorst von Wathlingen-Hänigsen. *Kali und Steinsalz*, No. **11**, 33–42 (1955).

These two papers describe the iron-analogue of boracite, found in several potassium salt deposits in the southern Harz in halite-anhydrite and halite-anhydrite-kieserite rock. The crystals, up to 4 mm. edge-length, range in color from light-green to raspberry-red and black. The pseudo-cubic crystals have the form (100) predominant, with (111) less prominent. Some crystals are zoned.  $X$ -ray study of 6 samples gave unit cells (on cubic indexing) ranging from 12.04 Å for white material to 12.12 Å for black, and 12.15 Å for raspberry-red with FeO 35.26, MnO 1.79% (Heide); K. and S. give  $a_0$  8.53,  $b_0$  8.53,  $c_0$  12.12 Å for the Mg end-member,  $a_0$  8.53,  $b_0$  8.60,  $c_0$  12.15 for material with Fe 2.21%,  $a_0$  8.58,  $b_0$  8.65,  $c_0$  12.17 for material with Fe 27.97, Mn 0.35%.  $G=3.17$ –3.27. Birefringent,  $n$  about 1.75 (K. and S.)

The following analyses are given: 1. Raspberry-red, from Bischofferode (partial analysis recalculated):  $\text{B}_2\text{O}_3$  50.56, FeO 35.26, MnO 1.79% (Heide). 2. Red-violet from Hänigsen. Fe 27.97, Mg 2.82, Mn 0.35, Cl 7.96, Br 0.0 83% (K. and S.) 3. Brownish-violet from Bischofferode: FeO 35.22, MgO 6.71, MnO 2.32, Cl 8.10, insol. 0.36% (K. and S.)

When heated the red material turns bluish-green at 115° (Heide), at 320–330° C. (K. and S.), and the color change is reversible on cooling. The blue-green color deepens, then changes to black at 800–1000° C. A sample cooled from 1000° C. showed no change in  $x$ -ray powder pattern. Melting began at 1100° C. (Heide). The orthorhombic-isometric inversion of ericaite occurs at 310–315° (265° for boracite) (K. and S.).

The name ericaite is quoted by K. and S. from a paper by H. Werner, Das Boracitvorkommen im Salzstock von Wathlingen-Hänigsen. *Der Aufschluss*, **1**, 24–26 (1950). This has not been seen.

M. F.



# CORRESPONDENTS, FELLOWS AND MEMBERS OF THE MINERALOGICAL SOCIETY OF AMERICA†

## CORRESPONDENTS

- ESKOLA, PENTTI, Geological Institute of the University, Snellmanink 5, Helsinki, Finland  
SLÁVÍK, FRANTISEK, Albertov 6, Praha, Czechoslovakia  
SPENCER, L. J., 111 Albert Bridge Road, London, S. W. 11, England  
TILLEY, C. E., Cambridge University, Cambridge, England

## FELLOWS AND MEMBERS

(\*Indicates Fellow)

- ABBAD, MANUEL, Instituto Fisica, Conzejo Superior de Investigaciones Cientificas, Serrano 119, Madrid, Spain  
ABELSON, PHILIP H., Geophysical Laboratory, 2801 Upton St., N. W., Washington 8, D. C.  
ADAMS, JOHN WAGSTAFF, U. S. Geological Survey, Denver Federal Center, Denver, Colorado  
ADLER, HANS H., 4401 Falcon St., Rockville, Maryland  
\*AGRELL, S. O., Dept. of Mineralogy, Downing St., Cambridge, England  
\*AHLFELD, FERERICO, Casilla 582, Cochabamba, Bolivia  
\*AHRENS, LOUIS H., Dept. of Geology and Mineralogy, University Museum, Oxford, England  
ALBANESE, JOHN S., P.O. Box 221, Union, New Jersey  
ALBEE, ARDEN L., 2217 Vernon Drive, Golden, Colorado  
ALBERS, JOHN P., 3728 King St., Alexandria, Virginia  
ALBRECHT, HERBERT O., 62 S. Hillcrest Rd., Springfield, Pennsylvania  
\*ALDRICH, HENRY R., 200 Van Orden Ave., Leonia, New Jersey  
ALESSIO, ORESTE G., JR., 908 Clinton Ave., Oak Park, Illinois  
ALEXANDER, A. E., 155 East 47th St., New York, N. Y.  
ALF, RAYMOND M., Webb School, Claremont, California  
ALLARD, GILLES, Chibougaman, Province Quebec, Canada  
ALLEN, ALFRED W., Dept. of Ceramic Engineering, University of Illinois, Urbana, Illinois  
ALLEN, JAMES, 322 West 23d St., South Sioux City, Nebraska  
ALLEN, ROBERT D., 332 Villanova Drive, Claremont, California  
\*ALLEN, VICTOR T., Institute of Geophysical Technology, 3621 Olive St., St. Louis 8, Missouri  
ALLEN, WALTER C., U. S. Steel Corporation, Research Laboratory, Kearny, New Jersey  
\*ALLING, HAROLD L., 155 West Jefferson Rd., Pittsford, N. Y.  
ALTSCHULER, Z. S., U. S. Geological Survey, Geochemistry & Petrology, Washington 25, D. C.  
ALVES DA SILVA, C., Sarmiento 72, Concepción, San Juan, Argentina  
\*AMOROS, JOSE L., Valencia 278, Barcelona, Spain  
AMPAGOOMIAN, SARKIS G., U. S. Bureau of Mines, College Park, Maryland  
AMSTUTZ, G. C., Pappelweg 32, Wabern-Bern, Switzerland  
ANDERSON, A. BENTON, Hazardville, Connecticut  
\*ANDERSON, ALFRED L., Dept. of Geology, Cornell University, Ithaca, New York  
ANDERSON, BARBARA J., P.O. Box 197, Rutherford, New Jersey  
ANDERSON, B. W., 55 Hatton Garden, London E. C., England

† Changes received by the Secretary after January 15, 1956, are not included.



- \*ANDERSON, CHARLES A., U. S. Geological Survey, Washington 25, D. C.
- \*ANDERSON, GEORGE H., 1931 RCA Building, 30 Rockefeller Plaza, New York 20, N. Y.
- \*ANDREATTA, CIRO, Istituto di Mineralogia e Petrografia, Università di Bologna, Piazza San Donato 1, Bologna, Italia
- ANDREW, GERALD, M/Y Dyfi, c/o C. J. Broom and Sons, Brundall, Norfolk, England
- ANDREWS, T. G., Geology Department, Box 2005, University, Alabama
- APPLEMAN, DANIEL E., Dept. of Geology, Johns Hopkins University, Baltimore 18, Maryland
- ARENSBURG, GUILLERMO, Casilla 9343, Santiago, Chile
- ARNOTT, RONALD J., Box 425, Bellaire, Texas
- ASHBY, GEORGE E., 12714 Feldon St., Silver Spring, Maryland
- AUSKERN, ALLAN, 1105 East 18th St., Brooklyn 30, New York
- AXELROD, JOSEPH, U. S. Geological Survey, Washington 25, D. C.
- AYE, TIN, Dept. of Geology, University of Wisconsin, Madison, Wisconsin
- AZAROFF, LEONID V., Armour Research Foundation, Technology Center, Chicago 16, Illinois
- BACON, CHARLES S., JR., Dept. of Geology & Mineralogy, Case Institute of Technology, Cleveland 6, Ohio
- BACON, JAMES F., Development & Research Dept., Corning Glass Works, Corning, New York
- \*BAILEY, EDGAR H., 257 Oakdale Way, Redwood City, California
- BAILEY, ROY A., U. S. Geological Survey, Washington 25, D. C.
- BAILEY, S. W., Dept. of Geology, University of Wisconsin, Madison 6, Wisconsin
- \*BAIN, GEORGE W., 22 Hitchcock Rd., Amherst, Massachusetts
- BAKER, W. S. A., c/o Mr. Rowlands, London Tin Corp. Ltd., 55-61 Moorgate, London EC 2, England
- \*BANDY, MARK C., Redfield, Iowa
- BANK, WALTER, 1406 4th St., Sparks, Nevada
- \*BANNERMAN, HAROLD M., U. S. Geological Survey, Washington 25, D. C.
- \*BANNISTER, F. A., 34 Monahan Ave., Purley, Surrey, England
- \*BARKSDALE, J. D., Dept. of Geology, University of Washington, Seattle 5, Washington
- \*BARLETT, HELEN BLAIR, AC Spark Plug Division, General Motors Corporation, Flint 2, Michigan
- \*BARNES, VIRGIL E., 207 East 33d St., Austin 21, Texas
- BARNES, WILLIAM C., Box 413, Golden, Colorado
- \*BARNES, WILLIAM H., Physics Division, National Research Council, Ottawa, Canada
- BARRETT, RICHARD L., 455 El Prado, Las Cruces, New Mexico
- \*BARTH, TOM. F. W., Mineralogisk Geologisk Museum, Oslo 45, Norway
- BARTHOLOMÉ, PAUL, Dept. of Geology, Princeton University, Princeton, New Jersey
- BASSETT, ALLEN M., 1450 Indian Hill Blvd., Claremont, California
- \*BATEMAN, ALAN M., Yale University, New Haven, Connecticut
- \*BATES, THOMAS F., Dept. of Earth Sciences, Pennsylvania State University, University Park, Pennsylvania
- BAUMANN, H. N., Research Dept., The Carborundum Company, Niagara Falls, New York
- BAXTER, E. F., Calle Viamonte 470, Corboda, Argentina
- BAUMGOLD, JOSEPH, 62 West 47th St., New York 26, New York
- BAWDEN, F. C. MICHELL, Geology Dept., New Consolidated Goldfields, Box 1167, Johannesburg, South Africa
- \*BECK, CARL W., Dept. of Geology, Indiana University, Bloomington, Indiana

- BECK, WARREN R., Minnesota Mining & Mfg. Co., 2301 Hudson Rd., St. Paul 6, Minnesota
- \*BEHRE, CHARLES E., JR., Dept. of Geology, Columbia University, New York 27, New York
- BELDEN, JOAN S., 3437 East Bunell St., Tucson, Arizona
- BELL, GORDON L., Dept. of Geology, University of North Dakota, Grand Forks, North Dakota
- \*BELL, JAMES F., 304 Public Service Bldg., Portland 4, Oregon
- BENHAM, MILFORD J., Phoenix College, Phoenix, Arizona
- BERMAN, JOSEPH, U. S. Bureau of Mines, College Park, Maryland
- BERMAN, ROBERT, 34 Prentiss St., Cambridge 38, Massachusetts
- BERNHEIMER, ALAN W., Dept. of Bacteriology, N.Y.U. College of Medicine, 550-1st Ave., New York 16, New York
- \*BERRY, LEONARD G., Dept. of Mineralogy, Queen's University, Kingston, Ontario, Canada
- BERTHOLD, SARAH M., U. S. Geological Survey, Washington 25, D. C.
- BESSON, WILLIAM W., 6 Byrd St., Rix Park, Rye, New York
- BETHKE, PHILIP M., Dept. of Geology, Missouri School of Mines, Rolla, Missouri
- BEVER, JAMES E., Geology Dept., Miami University, Oxford, Ohio
- BILGRAMI, S. A., Dept. of Geology, The University, Manchester 13, England
- \*BILLINGS, MARLAND P., Harvard University, Cambridge 38, Massachusetts
- BINGHAM, E. BURRIS, 8436 Ramsgate, Los Angeles 45, California
- BIRCHMAN, PAUL, 717 East Randolph St., Angola, Indiana
- BIREN, HELEN A. (Mrs.), 138-10 Cronston Ave., Belle Harbor, New York
- BJAREBY, GUNNAR, 147 Worthington St., Boston 15, Massachusetts
- BLADE, LAWRENCE V., 8194 Robinson Way, Arvada, Colorado
- BLANK, HORACE R., Dept. of Geology, Texas A. & M. College, College Station, Texas
- BLAYNEY, IRWIN B., Box 5366, Phoenix, Arizona
- BLONDEL, FERNAND A., 12 Rue de Bourgogne, Paris 7, France
- BLOSS, F. DONALD, Dept. of Geology, University of Tennessee, Knoxville, Tennessee
- BLYE, HAROLD, 93-14 240 St., Bellerose 26, Long Island, New York
- BOASE, WOODY CARL, A/2C Wilmer G. Boase, AF 15463009, 1st AFDS, APO 194, c/PM, New York, N. Y.
- BODELSEN, OSCAR W., 219 East Main St., Mount Kisco, New York
- BOGUE, RICHARD G., 2943 E. 21st St., Tucson, Arizona
- BONATTI, STEPHANO, Instituto di Mineralogia, Universita, Via Volta 2, Pisa, Italy
- \*BONORINO, FÉLIX GONZÁLEZ, Dept. of Geology, Missouri School of Mines, Rolla, Missouri
- BOOS, MARGARET F., 2036 South Columbine St., Denver 10, Colorado
- BOOTH, OLIVER E., 907 Clinton Ave., Des Moines 13, Iowa
- BOTINELLY, THEODORE, U. S. Geological Survey, Box 360, Grand Junction, Colorado
- BOTNER, CLEMENTS A., 331 Olive St., South, Orange, California
- BOUCOT, ARTHUR J., Room 334, U. S. National Museum, Washington 25, D. C.
- \*BOWEN, NORMAN L., Geophysical Laboratory, 2801 Upton St., N.W., Washington 8, D. C.
- BOWIE, STANLEY H. U., Geological Survey and Museum, Exhibition Rd., London S.W. 7, England
- \*BOWLES, OLIVER, 5000 Massachusetts Ave., N. W., Washington 16, D. C.
- BOYD, FRANCIS R., JR., Geophysical Laboratory, 2801 Upton St., N.W., Washington 8, D. C.
- BRADLEY, J. W., 4639 Crenshaw Boulevard, Los Angeles 43, California
- \*BRADLEY, WILLIAM F., Illinois State Geological Survey, Urbana, Illinois

- \*BRAMLETTE, MILTON N., Scripps Institute of Oceanography, La Jolla, California  
BRANT, ARTHUR M., 156 Hampden Park, Tiffin, Ohio  
BRECKE, ERVIN A., Rosiclare, Illinois  
BRENDLER, W., Bergkoppelweg 30 pt., Hamburg-Fuhlsbittel, Germany  
\*BRINDLEY, GEORGE W., Mineral Sciences Building, Pennsylvania State University, University Park, Pennsylvania  
BRINEMAN, JOHN H., 2800 Northwest Highway, Dallas 9, Texas  
BRISON, R. J., 2240 Zollinger Road, Columbus 21, Ohio  
BRITTON, MARVIN G., 139 Steuben St., Painted Post, New York  
BROBST, DONALD A., U. S. Geological Survey, Denver Federal Center, Denver, Colorado  
BROPHY, GERALD P., 88 Morningside Drive, New York 27, New York  
BROWN, GEORGE, Rothamstead Experiment Station, Harpenden, Hertzs, England  
BROWN, GEORGE MALCOLM, Dept. of Geology, Oxford University, Oxford, England  
BROWN, H. Trueheart, Geochemical Survey, 3806 Cedar Springs, Dallas, Texas  
\*BROWN, JOHN S., St. Joseph Lead Co., Bonne Terre, Missouri  
BROWN, W. R., Dept. of Geology, University of Kentucky, Lexington 29, Kentucky  
BROWNELL, WAYNE E., 126 Hornell St., Hornell, New York  
BRUNK, C. C., 4019 Leeward Ave., Los Angeles 5, California  
BRUNTON, GEORGE, Shell Development Co., 3737 Bellaire Blvd., Houston 25, Texas  
\*BRYANT, W. M. D., 703 Silverside Rd., Wilmington, Delaware  
BUCK, ALAN D., P.O. Drawer 2131, Jackson, Mississippi  
BUCK, DANIEL CLIFFORD, Westinghouse Electric Corporation, Elmira, New York  
BUCKLEW, HAROLD C., 24 Lyons Ave., Newark 8, New Jersey  
\*BUCKLEY, HAROLD E., 100 Hazelhurst Road, Worsley, Manchester, England  
\*BUDDINGTON, ARTHUR F., Dept. of Geology, Princeton University, Princeton, New Jersey  
\*BUERGER, MARTIN J., Dept. of Geology, Massachusetts Institute of Technology, Cambridge 39, Massachusetts  
\*BUERGER, NEWTON W., Naval Post Graduate School, Monterey, California  
\*BULLARD, FRED W., Dept. of Geology, University of Texas, Austin, Texas  
BULLOCK, KENNETH C., Dept. of Geology, Brigham Young University, Provo, Utah  
BÜLTEMANN, HANS W., Bahnhofstrasse 256, Bovenden über Göttingen, British Zone, Germany  
BUNDY, WAYNE M., Indiana Geological Survey, Indiana University, Bloomington, Indiana  
BUNTON, PAUL B., 2900 Connecticut Ave., N.W., Washington 8, D. C.  
\*BURBANK, WILBUR S., 3346 Runnymede Place, N.W., Washington 15, D. C.  
\*BURFOOT, J. DABNEY, JR., Dept. of Geology, Cornell University, Ithaca, New York  
BURGESS, DAVID A., P.O. Box 6667, Loiza Station, Santurce, Puerto Rico  
BURLEY, GORDON, National Bureau of Standards, Washington 25, D. C.  
BURNHAM, C. WAYNE, Dept. of Geology, Pennsylvania State University, University Park, Pennsylvania  
BURNHAM, G. E., 128 S. Encinitas Ave., Monrovia, California  
\*BURRI, CONRAD, Mineralogisch-Petrographisches Institut, Eidgenössische Technische Hochschule, Zürich 6, Switzerland  
BURST, JOHN F., 3737 Bellaire Blvd., Shell Oil Company, Houston, Texas  
\*BUTLER, G. MONTAGUE, 1147 South Van Ness Ave., Santa Ana, California  
BUTLER, JOHN W., JR., c/o Phillips Venezuelan Oil Company, Apartado 1031, Caracas, Venezuela  
BUTLER, KEITH H., Sylvania Electric Products, Inc., 60 Boston Street, Salem, Massachusetts  
\*BUTLER, ROBERT D., Caixa Postal 396, Belem do Para, Brazil

- \*BUTTGEBACH, HENRY J., Avenue Madoux 129, Woluwe St. Pierre, Belgium  
BYERS, FRANK M., JR., 120 Texas Avenue, Grand Junction, Colorado  
BYRON, J. E., 534 Highland Ave., Boulder, Colorado
- CALLAHAN, WILLIAM H., 1 Rowe Place, Franklin, New Jersey  
CALLAWAY, A. L., Box 258, Lakeland, Florida  
CALVER, JAMES L., 1553 Cristobal Drive, Tallahassee, Florida  
CALVERT, EARL L., 8844 Ardendale Ave., San Gabriel, California  
CAME, JOHN T., 4059 45 St., San Diego 5, California  
\*CAMERON, EUGENE N., Science Hall, University of Wisconsin, Madison 6, Wisconsin  
CAMERON, THOMAS B., JR., Dept. of Chemistry, University of Cincinnati 21, Ohio  
\*CAMPBELL, CHARLES D., Route 1, Pullman, Washington  
CAMPBELL, DOUGLAS D., Eldorado P.O., Saskatchewan, Canada  
\*CAMPBELL, IAN, California Institute of Technology, Pasadena 4, California  
\*CANNON, RALPH S., JR., U. S. Geological Survey, Denver Federal Center, Denver, Colorado
- CARDONA, LUIS E., 120 West 68th St., New York 25, New York  
CARGILLE, R. P., 118 Liberty St., New York 6, New York  
CARHART, GRACE M., 112 Main Street, Peekskill, New York  
CARL, HOWARD P., Highland, Howard County, Maryland  
\*CAROBBI, GUIDO, Istituto di Mineralogia della Università, Florence, Italy  
CARROLL, DOROTHY, U. S. Geological Survey, Washington 25, D. C.  
CARRON, MAXWELL K., U. S. Geological Survey, Washington 25, D. C.  
CARTRALHO DA SILVA, JAIR, Escola de Minas, Ouro Preto, Minas Gerais, Brazil  
CASPERSON, WILLIAM C., Paterson Museum, 268 Summer St., Paterson, New Jersey  
CHACE, FRED M., 3950 Langley Court, N.W., Washington 16, D. C.  
CHANG, KUAN-HUAN, 5559 S. Blackstone Ave., Chicago 37, Illinois  
CHANG, PING HSI, c/o Arthur Montgomery, 277 Park Ave., New York 17, New York  
\*CHAPMAN, CARLETON A., Dept. of Geology, University of Illinois, Urbana, Illinois  
\*CHAPMAN, RANDOLPH, Dept. of Geology, Trinity College, Hartford, Connecticut  
CHASE, GERALD W., Oklahoma Geological Survey, Norman, Oklahoma  
\*CHAYES, FELIX, Geophysical Laboratory, 2801 Upton St., N.W., Washington 8, D. C.  
CHESLEY, FRANK G., Central Research Laboratories, Inc., Red Wing, Minnesota  
\*CHESTERMAN, CHARLES W., 1066 Keith Ave., Berkeley, California  
CHIDESTER, ALFRED H., 160½ Main St., Montpelier, Vermont  
\*CHRIST, CHARLES L., Geochemistry and Petrology Branch, U. S. Geological Survey, Washington 25, D. C.
- CHRISTMAN, ROBERT A., Dept. of Geology, Cornell University, Ithaca, New York  
CHROMY, BENJAMIN J., 301 Westory Building, Washington 5, D. C.  
CHUDOBA, KARL F., Angerstrasse 263, Adelehsen bei Göttingen (20b), Germany  
CHUTE, NEWTON E., Dept. of Geology, Syracuse University, Syracuse, New York  
\*CLABAUGH, STEPHEN E., Dept. of Geology, University of Texas, Austin 12, Texas  
CLAFFY, ESTHER W. (Mrs.), Naval Research Laboratory, Code 6440, Luminescence Section, Washington 25, D. C.  
\*CLARINGBULL, G. F., Dept. of Mineralogy, British Museum (Natural History), Cromwell Road, London, S.W. 7, England
- CLARK, JOAN R., 3 Pook's Hill Road, Bethesda 14, Maryland  
CLAUSEN, HANS, Mineralogical Museum, Oster Voldgade 7, Copenhagen, K., Denmark  
CLEMENS, LELAND P., 8304 Bevan St., San Gabriel, California  
CLINE, STANLEY H., North Road, MC 57, Warren, Ohio

- \*COATS, ROBERT, U. S. Geological Survey, Washington 25, D. C.  
COCKBAIN, ALAN GRAY, St. John's College, Cambridge, England  
COES, LORING, JR., Fiskdal Rd., Brookfield, Massachusetts  
COHEN, ALVIN J., Mellon Institute, 4400 Fifth Ave., Pittsburgh 13, Pennsylvania  
\*COLBURN, BURNHAM S., Greystone Court, Biltmore, North Carolina  
\*COLBURN, WILLIAM B., P.O. Box 456, Rancho Santa Fe, California  
COLE, G. THURSTON, Rumford Point, Maine  
COLEMAN, LESLIE C., 354 Van Norman, Port Arthur, Ontario, Canada  
COLEMAN, ROBERT G., U. S. Geological Survey, Geochemistry and Petrology, Washington 25, D. C.  
COLLINI, BENGT, Rundelsgränd 6 D, Uppsala, Sweden  
COLLINS, BARBARA J., AO 3010221, 66th RECONTECHRON, APO 227, U. S. Army, New York, N. Y.  
COMEFORO, JAY EUGENE, 36 Seventh St., Frenchtown, New Jersey  
COOK, KATHRYN H. (Mrs.), 115 Firestone Blvd., Akron 1, Ohio  
COOK, WILLIAM R., JR., 1067 Hillstone Rd., Cleveland Heights 21, Ohio  
COOKE, H. R., JR., Martin, Sykes, Woods and Assoc., Apartado 2021, Caracas, Venezuela  
\*COOKE, S. R. B., Minnesota School of Mines, Minneapolis 14, Minnesota  
COOLEY, MARY E., 703 South Forest Ave., Ann Arbor, Michigan  
COOMBS, D. S., Geology Dept., University of Otago, Dunedin, N. 1, New Zealand  
COONEY, ROBERT L., 1828 So. Stoner, Los Angeles 25, California  
\*COOPER, JOHN R., U. S. Geological Survey, Denver Federal Center, Denver, Colorado  
\*CORBETT, CLIFTON S., 045 Tremont Ave., Westfield, New Jersey  
COREY, ALLAN F., 1760 North Eighth, Apt. 4, Grand Junction, Colorado  
\*CORRENS, CARL W., Lotzestrasse 13, Göttingen, Germany, British Zone  
CORWIN, GILBERT, Military Geology Branch, 29th Engr. Bn Base Topo, APO 500, San Francisco, California  
COTTER, PERRY G., 24 East Circle Rd., Norris, Tennessee  
COUDRAY, R. M., Star Route, ACM Box 53, Grants, New Mexico  
COX, JOSEPH E., JR., R.D. 2, Pottstown, Pennsylvania  
CRAWFORD, ARTHUR L. Utah Geol. Miner. Survey, Mines Bldg., University of Utah, Salt Lake City, Utah  
CREITZ, ELLIS E., Drawer L, University, Alabama  
CRIBBS, RICHARD E., 3812 Terrace View Rd., Knoxville 18, Tennessee  
CRIPPEN, RICHARD A., JR., Division of Mines, Ferry Building, San Francisco, California  
CROFT, WILLIAM J., Massachusetts Inst. of Technology, Lincoln Laboratory, Box 73, Lexington 73, Massachusetts  
CROOK, THEO H., 45 Alta Rd., Berkeley 8, California  
CRUMP, ROBERT M., Jones & Laughlin Steel Corp., Geol. Division, 401 Liberty Ave., Gateway Center, Pittsburgh, Pennsylvania  
CUERVO, Dra. America Ana, Laboratoria No. 1 de Mineralogia, Escuela de Ciencias, Universidad de la Havana, Havana, Cuba  
CUMMINS, A. B., Morning Glory Rd., Bound Brook, New Jersey  
\*CURRIER, LOUIS W., U. S. Geological Survey, Washington 25, D. C.  
CUTTITTA, FRANK, 12911 Bluehill Road, Silver Spring, Maryland  
  
DAGGETT, E. B., c/o Vanadium Corporation of America, Naturita, Colorado  
DAHL, HARRY M., AEC Geologic Branch, Box 270, Grand Junction, Colorado  
DAKE, H. C., 329 S. E. 32d Ave., Portland 15, Oregon  
DANNER, WILBERT R., Dept. of Geology and Geography, University of British Columbia, Vancouver 8, B. C., Canada



- DARNEAL, ROBERT L., Box 832, Stanford University, Stanford, California
- DAVES, DELMER LAWRENCE, 107 N. Bentley Ave., West Los Angeles 24, California
- DAVEY, W. P., Osmond Laboratory, Pennsylvania State University, University Park, Pennsylvania
- \*DAVIDSON, CHARLES F., Dept. of Geology, The University, St. Andrews, Scotland
- DAVIDSON, STANLEY C., 30 Elm Ave., Apt. 206, Toronto 5, Ontario, Canada
- DAVIS, DONALD W., RFD 1, Katonah, New York
- \*DAVIS, GORDON L., 2 Anita Court, Rockville, Maryland
- DAVIS, HARRY T., North Carolina State Museum, Raleigh, North Carolina
- \*DE BRETTEVILLE, A. P., JR., 224 Navesink Ave., Highlands, New Jersey
- DELARIO, A. J., 316 Broadway, Paterson, New Jersey
- DE LAS CASAS, FERNANDO, Alcanfores 125, Miraflores-Lima, Peru
- DENGO, GABRIEL, Apartado 1851, San Jose, Costa Rica, Central America
- DENNING, REYNOLDS M., Mineralogical Laboratory, University of Michigan, Ann Arbor, Michigan
- DENNY, M. V., Electrotechnical Laboratory, U. S. Bureau of Mines, Norris, Tennessee
- DESANTIS, HELEN V. E. (Mrs.), 86-16 60th Rd., Elmhurst, Long Island, New York
- DESMOND, THOMAS C., 94 Broadwav, Newburgh, New York
- DETTMER, FRIEDRICH, Herderstrasse 22, Box 620, Lübeck, Germany
- DEUDON, M. (Mlle.), Institut de Recherches de la Siderurgie, Service Minerais, 6 rue de Lota, Paris XVI, France
- DEUL, MAURICE, U. S. Geological Survey, Geochemistry & Petrology, Washington 25, D. C.
- \*DEVILLIERS, JACOB E., Geology Dept., University of Orange Free State, Bloemfontein, South Africa
- DEVORE, GEORGE W., Dept. of Geology, University of Chicago, Chicago 37, Illinois
- DEVRIES, ROBERT C. 2012 Hudson St., Schenectady, New York
- DIETERICH, J. F., 11144 Riverside Drive, North Hollywood, California
- DIETZ, RALPH W., 106 B Bard, China Lake, California
- DIETZ, ROBERT SINCLAIR, ONRL (Box 39), Navy 100, c/o Fleet P.O., New York, New York
- DILL, ROBERT, 846 College St., Beloit, Wisconsin
- DODGE, NELSON B., Applied Research Laboratory, U. S. Steel Corporation, Monroeville, Pennsylvania
- \*DODGE, THEODORE A., 635 North Third Ave., Tucson, Arizona
- DOEGLAS, D. J., Laboratorium voor Mineralogie en Geologie, Duivendaal 2, Wageningen, Holland
- \*DOLAR-MANTUANI, LUDMILA M. M., Hydro-Electric Power Commission Laboratory, 620 University Avenue, Toronto 2, Canada
- \*DONNAY, GABRIELLE (Mrs.), c/o Chemistry Dept., Johns Hopkins University, Baltimore 18, Maryland
- \*DONNAY, JOSEPH D. H., Dept. of Geology, Johns Hopkins University, Baltimore 18, Maryland
- DORNE, ARTHUR, 30 Sylvester St., Westbury, New York
- DOSSE, A. F., 8805 Hemlock Ave., Fontana, California
- DOTEN, ROBERT K., Dept. of Geology, University of Vermont, Burlington, Vermont
- DOUGLASS, ROBERT M., 3785 Gold St., Apt. 3, Los Alamos, New Mexico
- DRAEGER, R. H. (Capt.), Medical Research Dept., Mine Countermeasures Station, Panama City, Florida
- DRAGSDORF, R. DEAN, Dept. of Physics, Kansas State College, Manhattan, Kansas
- DRAISIN, WILBURT M., 23 Pierrepont Rd., Newton Lower Falls 62, Massachusetts

- \*DREYER, ROBERT M., Exploration Dept., Kaiser Aluminum and Chemical Corp., 1924 Broadway, Oakland 12, California  
DRUMMOND, PAUL L., 1203 Bartlett, Apt. 7, Houston 6, Texas  
DRURY, WILBERT C., Arthur, Ontario, Canada  
DUDAS, LASZLO, 860 Victory Village, Henderson, Nevada  
DUERSMITH, L. J., 405 Poplar St., Columbia, Pennsylvania  
DUFFENDACK, O. S., Philips Laboratories, Inc., Irvington-on-Hudson, New York  
DUHOVNIK, J. M., Dept. of Mineralogy, University, Ljubljana, Yugoslavia  
DUPLIN, V. J., Jr., 300 North Ave., Fanwood, New Jersey  
DuPONT, JAMES M., Thermoplastic, Inc., Stirling, New Jersey  
DWORNIK, EDWARD J., 4611 62d Ave., Landover, Maryland
- \*EARLEY, J. W., P. O. Drawer 2038, Pittsburgh 30, Pennsylvania
- \*EBBUTT, FRANK, 173 Glenrose Ave., Toronto 5, Canada  
EBERLEIN, G. DONALD, Box 372 Los Gatos, California  
EBERT, HEINZ, Avenida Pasteur 404, Commissao Nacional da Producao Mineral, Rio de Janeiro, Brazil
- \*ECKEL, EDWIN B., U. S. Geological Survey, Denver Federal Center, Denver, Colorado  
ECKELMANN, F. DONALD, 607 Schermerhorn, Columbia University, New York 27, New York
- \*ECKERMANN, HARRY VON, Edeby, Sparreholm, Sweden  
ECKERT, H. W., 110 East Main St., Florence, Colorado  
EDDY, C. N., National State Bank, Boulder, Colorado
- \*EDWARDS, AUSTIN B., Geology Dept., University of Melbourne, Melbourne, Australia  
EFREMOV, NICHOLAS, 1061 Intervale Ave., Bronx, New York, New York  
EHLERS, ERNEST G. Dept. of Mineralogy, Lord Hall 140, Ohio State University, Columbus 10, Ohio  
EHRMANN, MARTIN L., c/o S. Ehrmann, 1961 N. Argyle Ave., Los Angeles 28, California  
EISENBERG, MARVIN, Armour Research Foundation, Technology Center, Chicago 16, Illinois
- \*EITEL, WILHELM H. J., Director of Silicate Institute, University of Toledo, Toledo 6, Ohio  
ELDER, CURTIS H., 4125 East 31 St., Tulsa, Oklahoma
- \*ELLESTAD, REUBEN B., 712 South Cedar Lake Rd., Minneapolis 5, Minnesota  
ELWELL, WILBUR J., 2 Duck St., Danbury, Connecticut  
EMBREY, PETER G., Dept. of Mineralogy, University Museum, Oxford, England  
EMERSON, DONALD O., 116 Mineral Industries Bldg., Pennsylvania State University, University Park, Pennsylvania
- \*EMMONS, RICHARD C., Science Hall, University of Wisconsin, Madison, Wisconsin
- \*ENGEL, ALBERT E. J., Dept. of Geology, California Institute of Technology, Pasadena 4, California  
ENGEL, RENE, 1222 Blair Ave., South Pasadena, California  
ENLOWS, HAROLD E., University of Tulsa, Seventh and College, Tulsa 4, Oklahoma  
ENZMANN, ROBERT D., 4800 Lincoln Place Drive, Des Moines, Iowa  
ERD, RICHARD C. Geochemistry & Petrology, U. S. Geological Survey, Washington 25, D. C.  
ERDMANN, CHARLES E., U. S. Geological Survey, P.O. Box 1827, Great Falls, Montana
- \*ERICKSON, EDWIN S., JR., Dept. of Mineralogy, Pennsylvania State University, University Park, Pennsylvania  
ERICKSON, GEORGE E., U. S. Geological Survey, Foreign Branch, Washington 25, D. C.

- ERNST, T., Mineralogisches Institut der Universität Erlangen, Schlossgarten 5, Erlangen, Germany
- EVANS, HAROLD, Box 425, Lower State Rd., Doylestown, Pennsylvania
- \*EVANS, HOWARD T., JR., Geochemistry & Petrology Branch, U. S. Geological Survey, Washington 25, D. C.
- FACKLER, WILLIAM C., Rt. 3, Box 509, Golden, Colorado
- \*FAESSLER, CARL, Université Laval, Boulevard de l'Entente, Quebec, Canada
- \*FAHEY, JOSEPH J., U. S. Geological Survey, Washington 25, D. C.
- \*FAIRBAIRN, HAROLD W., Dept. of Geology, Massachusetts Institute of Technology, Cambridge 39, Massachusetts
- \*FANKUCHEN, I., Polytechnic Institute of Brooklyn, Brooklyn, New York
- FARWELL, FRED W., 6 Pasture Lane, Norton, Connecticut
- \*FAUST, GEORGE T., 9907 Capitol View Ave., Silver Spring, Maryland
- FEINSTEIN, HYMAN I., 1400 Drexel St., Takoma Park 12, Maryland
- \*FERGUSON, R. B., Dept. of Geology, University of Manitoba, Winnipeg, Canada
- \*FETTKE, CHARLES R., 1118 Wightman St., Squirrel Hill Station, Pittsburgh 17, Pennsylvania
- FILER, ALVIN V., JR., 1447 Alford Ave., Birmingham 9, Alabama
- FILER, RUSSELL, 1344 Highway 99, San Bernardino, California
- FINCH, JAMES, Geology School, University of Melbourne, Carlton N 3, Victoria, Australia
- FINLAY, G. R., c/o Norton Company, Chippawa, Ontario, Canada
- FINNEGAN, MICHAEL J., Box 1127, Silsbee, Texas
- \*FISHER, D. JEROME, Dept. of Geology, University of Chicago, Chicago 37, Illinois
- FISHER, ROBERT B., 101 West 14th, Rolla, Missouri
- FISK, HENRY G., University of Wyoming, Laramie, Wyoming
- FLACK, WYLIE H., 333 Borbeck St., Philadelphia 11, Pennsylvania
- FLAGG, A. L., Box 2345, Phoenix, Arizona
- FLEISCHER, ARTHUR, Box 549, Easthampton, Massachusetts
- \*FLEISCHER, MICHAEL, U. S. Geological Survey, Washington 25, D. C.
- FOLCH, JOAQUIN, Calle Provenza 302, Barcelona, Spain
- FOLDVART, ALADAR, Hungarian Geological Survey, Stefania St. 14, Budapest 14, Hungary
- FONTALTABE, MANUEL, Rocafort, III, 2º2ª, Barcelona, Spain
- \*FORD, HUGH ALEXANDER, 736 Riverside Drive, Apt. 6-C, New York 31, New York
- FORMAN, S. A., Physical and Crystal Chemistry Section, Mines Branch, Ottawa, Ontario, Canada
- FORSLEV, ALBERT W., Dept. of Geology, University of Chicago, Chicago 37, Illinois
- \*FOSHAG, WILLIAM F., U. S. National Museum, Washington 25, D. C.
- \*FOSTER, MARGARET D., U. S. Geological Survey, Washington 25, D. C.
- FOSTER, PHILIP C., R. D. 1, South Berwick, Maine
- \*FOSTER, WILFRID R., Dept. of Mineralogy, Ohio State University, Columbus 10, Ohio
- FOSTER, WILLIAM J., 511 Madison Place, Falls Church, Virginia
- FOUCHE, JEAN, 324 Rebecca St., Pretoria, South Africa
- FOX, PORTLAND P., Cherokee Gardens, Cleveland, Tennessee
- \*FRANCO, RUI RIBEIRO, Universidade de Sao Paulo, Caixa Postal 8105, Sao Paulo, SP Brazil
- FRANK, JACOB N., Geology Dept., Creole Petroleum Corp., Apartado 889, Caracas, Venezuela
- \*FRASER, DONALD M., Geological Dept., Bethlehem Steel Company, Bethlehem, Pennsylvania

- \*FRASER, HORACE J., Room 1506, 44 King St. West, Toronto, Canada  
FRÉCHETTE, V. D., N. Y. State College of Ceramics, Alfred, New York  
FREEZE, ARTHUR C., 52 First Ave., Chapman Camp, British Columbia, Canada  
FRENCH, BEVAN, 98 Alexander Ave., Nutley 10, New Jersey  
FRENCH, GORDON B., Geology Dept., Missouri School of Mines, Rolla, Missouri  
FRETZ, A. HENRY, 42 West Market St., Bethlehem, Pennsylvania  
FRIEDMAN, G. L., Box 288, Woodland Hills, California  
FRIEDMAN, GERALD M., 814 Wellington E., Sault Ste. Marie, Ontario, Canada  
FRIES, CARL, JR., U. S. Geological Survey, Washington 25, D. C.  
FRITTS, JOHN J., Box 25, Metaline Falls, Washington  
\*FRONDEL, CLIFFORD, 12 Geological Museum, Oxford St., Cambridge 38, Massachusetts  
FRONDEL, JUDITH W. (Mrs.), Beatrice Circle, Belmont, Massachusetts  
\*FRUEH, ALFRED J., JR., Dept. of Geology, University of Chicago, Chicago 37, Illinois  
FUJII, TAKASHI, Institute of Silicate Research, University of Toledo, Toledo 6, Ohio  
\*FULLER, RICHARD E., 3801 East Prospect St., Seattle 2, Washington  
FUOG, H. L., 4149 Barrett Rd., Los Angeles 32, California  
FURBISH, WILLIAM J., Box 6665 College Station, Duke University, Durham, North Carolina  
\*FURNIVAL, GEORGE M., c/o The California Standard Co., 200 Greyhound Bldg., Calgary, Alberta, Canada  
FUSTER, JOSE M., Laboratorios de Geologia, Universidad Central, San Bernardo 51, Madrid, Spain  
FYFE, WILLIAM S., Chemistry Dept., University of Otago, Dunedin, New Zealand  
  
\*GALBRAITH, F. W., Dept. of Geology, University of Arizona, Tucson, Arizona  
\*GALLAGHER, DAVID, U. S. Geological Survey, 4 Homewood Place, Menlo Park, California  
\*GALLITELLI, PAOLO, Istituto di Mineralogia della Università, Modena, Italy  
GALLONI, ERNESTO, Yerbal 1763, Buenos Aires, Argentina  
\*GARRELS, ROBERT M., Dept. of Geology, Harvard University, Cambridge 38, Massachusetts  
GATES, ROBERT M., Science Hall, Madison, Wisconsin  
\*GAUDIN, A. M., Room 8-211, Massachusetts Institute of Technology, Cambridge 39, Massachusetts  
\*GAULT, H. RICHARD, Dept. of Geology, Lehigh University, Bethlehem, Pennsylvania  
GAYLORD, GUILFORD B., Gabbs, Nevada  
GEORGALAS, GEORGE C., 39A Pipinou St., Athens, Greece  
GEORGE, D'ARCY R., 107 Spring Rd., Bedford, Massachusetts  
GERBER, KARL V., 11654 Forest Grove St., El Monte, California  
GHEITH, MOHAMED A., Dept. of Geology, Faculty of Science, Ein Shams University, Abbassia, Cairo, Egypt  
\*GIANELLA, VINCENT P., Mackay School of Mines, Reno, Nevada  
GIARDINI, A. A., 64 Blackman Rd., Grand Island, New York  
GIFFORD, JAMES C., 521 East Waverly St., Tucson 10, Arizona  
GILBERT, MARNA W., 2011 North Raymond Ave., Pasadena, California  
GILLESPIE, T. B., Star Route 2, St. Augustine Beach, St. Augustine, Florida  
GILLINGHAM, THOMAS E., 284 Morris Ave., Mountain Lakes, New Jersey  
\*GILLSON, JOSEPH L., 6017 du Pont Bldg., Wilmington 98, Delaware  
\*GILLULY, JAMES, U. S. Geological Survey, Denver Federal Center, Denver, Colorado  
GINDY, AMIN R., 27 Corniche (Block C), Chathy, Alexandria, Egypt  
GIRAULT, J. P., 16, St. Denis Ave., Quebec City, Quebec, Canada  
GIRHARD, M. NANCY, U. S. Geological Survey, Washington 25, D. C.



- GIVENS, DAVID B., Room 308, Masonic Bldg., Yakima, Washington
- GLACON, JULES, Service de la Carte Geologique, 14 Boulevard Baudin, Alger, Algeria
- GLASS, HERBERT D., Illinois State Geological Survey, Urbana, Illinois
- GERMAN, D. H., 69 Northdale Blvd., Toronto 13, Ontario
- \*GLASS, JEWELL J., U. S. Geological Survey, Washington 25, D. C.
- \*GODDARD, EDWIN N., Dept. of Geology, University of Michigan, Ann Arbor, Michigan
- \*GOLDICH, SAMUEL S., Dept. of Geology, University of Minnesota, Minneapolis 14, Minnesota
- \*GOLDSMITH, J. H., Dept. of Geology, University of Chicago, Chicago 37, Illinois
- \*GOODSPEED, GEORGE E., Dept. of Geology, University of Washington, Seattle, Washington
- GOUDEY, HATFIELD, Drawer M, Gabbs, Nevada
- GOWER, JOHN A., Kennco Explorations (Canada) Ltd., 25 King St. West, Toronto 1, Ontario, Canada
- GRAF, DONALD L., Illinois State Geological Survey, Urbana, Illinois
- GOWER, JOHN ARTHUR, 25 Westgate, Cambridge 38, Massachusetts
- GRAHAM, A. R., 49 Touraine Ave., Wilson Heights P.O., North York, Ontario, Canada
- \*GRAHAM, R. P. D., 775 Davaar Ave., Outremont, Quebec, Canada
- GRAHL, HARRY, 885 Cauldwell Ave., Bronx 56, New York
- \*GRATON, LOUIS C., Pleasant Hill Rd., Orange, Connecticut
- GRAVES, HOWARD B., JR., 826 South Ingraham Ave., Lakeland, Florida
- \*GRAW, OLIVER R., Dept. of Geology, Missouri School of Mines, Rolla, Missouri
- GREEN, ROBERT S., 18 South Jefferson Rd., Mexico, Missouri
- GREENBERG, SEYMOUR, Indiana Geological Survey, Bloomington, Indiana
- GREENBERG, SIDNEY A., Johns Manville Research Center, Manville, New Jersey
- \*GREENE, KENNETH T., 508 Remington St., Fort Collins, Colorado
- GREGORY, ROBERT F., 404 East Dexter, Covina, California
- \*GRIEG, JOSEPH W., Geophysical Laboratory, 2801 Upton St., N.W., Washington 8, D. C.
- \*GRIFFITHS, JOHN CEDRIC, Dept. of Earth Sciences, Pennsylvania State University, University Park, Pennsylvania
- GRIFFITHS, WALLACE R., U. S. Geological Survey, Washington 25, D. C.
- GRIGGS, ROY L., 1008 Montclair N.E., Albuquerque, New Mexico
- \*GRIM, RALPH E., 234 Natural History Bldg., University of Illinois, Urbana, Illinois
- GRIP, ERLAND, Boliden Mining Company, Boliden, Sweden
- GROGAN, ROBERT M., Room 6015, du Pont Bldg., Wilmington, Delaware
- GROSS, EUGENE B., 2705 Bonita, Grand Junction, Colorado
- GROSSMAN, IRVING G., Box 590, U. S. Geological Survey, Albany, New York
- \*GROUT, FRANK F., University of Minnesota, Minneapolis 14, Minnesota
- GRUNENFELDER, MARC, 9 Schlossgasse, Zürich, Switzerland
- \*GRUNER, JOHN W., University of Minnesota, Minneapolis 14, Minnesota
- GUDE, ARTHUR JAMES, III, Box 374, Golden, Colorado
- GUEST, H. J. M., Box 54, Klerksdorp, Transvaal, South Africa
- GUILLEMIN, CLAUDE, 6 rue Pierre Bourdan, Paris 12, France
- GUIMARAES, DJALMA, Rua Gonclaves Dias, 3182, Belo Horizonte, Brazil
- GULBRANDSEN, R. A., U. S. Geological Survey, 4 Homewood Place, Menlo Park, California
- \*GUMMER, WILFRID K., 201 Castner St., Arvida, Quebec, Canada
- GUNNELL, E. M., 3365 East Kentucky Ave., Denver 9, Colorado
- GUSH, I. N. (Mrs.), Box 1128, Johannesburg, South Africa
- GUTTORMSEN, PAUL A., JR., The Texas Co., Box 1346, Casper, Wyoming



- \*GYSIN, M. E., Laboratoire de Minéralogie de l'Université, Quai de l'Ecole-de-Médecin, Genève, Switzerland
- HADDOCK, WILLIAM J., JR., 23727 Lucille Ave., Torrance, California
- HADLEY, JARVIS B., General Geology Branch, U. S. Geological Survey, Denver Federal Center, Denver, Colorado
- \*HAFF, JOHN C., Dept. of Geology, Mount Holyoke College, South Hadley, Massachusetts
- HAGNER, ARTHUR F., Dept. of Geology, University of Illinois, Urbana, Illinois
- HAHN, THEODORE, Dept. of Geology, Room 24-007, Massachusetts Institute of Technology, Cambridge 39, Massachusetts
- HALBOUTY, M. T., Suite 729-32, Shell Building, Houston 2, Texas
- HALL, F. P., 206 Granger Rd., Syracuse 4, New York
- \*HALLIMOND, ARTHUR F., 75 Corringham Rd., London NW 11, England
- HALLOWELL, HORACE J., RFD 2, Ridgefield, Connecticut
- HALSEY, JONATHAN H., 101 Woodland Drive, Irving, Texas
- HAM, WILLIAM EUGENE, State Geological Survey, Norman, Oklahoma
- HAMILTON, PEGGY-KAY, 136 Park Ave. Bronxville 8, New York
- HANLEY, JOHN B., U.S.G.S. Building, A.R.C., Beltsville, Maryland
- HARADA, ZYUMPEI, Rigakubu-Chishitsu Kyoshitsu, Hokkaido Daigaku, Sapporo, Japan
- \*HARCOURT, GEORGE ALAN, Copper Cliff, Ontario, Canada
- HARDY, John A., RFD 1, Salisbury, Connecticut
- HARRIS, PETER G., Dept. of Geology, The University, Leeds 2, England
- HARRISON, JACK E., U. S. Geological Survey, GSA Building, Room 4232 Washington 25, D. C.
- HASEGAWA, SYUZO, c/o Cansekigaku-Kyoshitsu, Tohoku University, Sendai, Japan
- HASTINGS, EARL L., 318 Cherokee Trail, Lakeland, Florida
- \*HATCH, ROBERT A., 4135 North Bellevue Ave., White Bear Lake 10, Minnesota
- HATHAWAY, JOHN C., U. S. Geological Survey, Washington 25, D. C.
- HAUSEN, DONALD M., U. S. Bureau of Mines, 1605 Evans Ave., Reno, Nevada
- HAWKES, H. E., JR., Geology Dept., Massachusetts Institute of Technology, Cambridge 39, Massachusetts
- \*HAWLEY, JAMES E., Dept. of Geology, Queen's University, Kingston, Ontario, Canada
- HAYASE, KITARO, Rikogaku-bu-Kozangaku-Kyoshitsu, Totsuka, Shinjuku-ku, Tokyo, Japan
- \*HAYCOCK, MAURICE H., 552 Booth St., Ottawa, Canada
- HEARN, JANE K., 525 West Capitol St., Salt Lake City, Utah
- HEARNE, L. F., Echo Point, Wheeling, West Virginia
- HEATH, DONALD L., Ohio Brass Company, Barberton, Ohio
- HEINEMAN, ROBERT E. S., 20 Calle Conquista, Tucson, Arizona
- \*HEINRICH, F. WM., Mineralogical Laboratory, University of Michigan, Ann Arbor, Michigan
- HENDERSON, DONALD M., Dept. of Geology, University of Illinois, Urbana, Illinois
- \*HENDERSON, EDWARD P., U. S. National Museum, Washington 25, D. C.
- \*HENDRICKS, STERLING B., 1118 Dale Drive, Silver Spring, Maryland
- \*HENRY, NORMAN F. M., University of Cambridge, Downing St., Cambridge, England
- HENTSCHEL, H. W., 3100 DeKalb St., Washington Square, Norristown, Pennsylvania
- HERREID, GORDON, Box 34, College, Alaska
- HERRMANN, F., Postfach 20820, Basel 1, Switzerland
- HERZ, NORMAN, U. S. Geological Survey, 270 Dartmouth St., Boston 16, Massachusetts
- HERZENBERG, ROBERT, Casilla 23, Oruro, Bolivia, South America
- HESS, HAROLD D., Rt. 4, Box 177, Corvallis, Oregon
- \*HESS, HARRY H., Dept. of Geology, Princeton University, Princeton, New Jersey

- HEYL, ALLEN V., JR., U. S. Geological Survey, Mineral Deposits Branch, Agricultural Research Center, Beltsville, Maryland
- \*HEWETT, DONNEL F., 1460 Rose Villa St., Pasadena, California
- \*HEYL, GEORGE R., Apartado 889, Creole Petroleum Corporation, Caracas, Venezuela
- \*HIETANEN-MAKELA, ANNA M., U. S. Geological Survey, 4 Homewood Place, Menlo Park, California
- \*HIGAZY, RIAD A., Geology Dept., Faculty of Science, University of Alexandria, Moharram Bey, Alexandria, Egypt
- HILDEBRAND, FRED A., 9921 Dickens Ave., Bethesda, Maryland
- HILL, H. STANTON, Geology Dept., Pasadena City College, Pasadena 4, California
- HILL, W. G., Dept. of Geological Sciences, University of Toronto, Toronto, Ontario, Canada
- HILL, WILLIAM L., 10504 Royal Rd., Silver Spring, Maryland
- HILLS, RALPH F., 18 Greenwood Ave., Hartford 12, Connecticut
- HILLY, J., 6 Passage du Commandant Susini, Alger, Algeria
- HILMY, MOHAMED E., Dept. of Geology, Faculty of Science, Moharram Bey, Alexandria, Egypt
- HINCKLEY, ARTHUR T., 937 Harrison Ave., Niagara Falls, New York
- HINZ, GEORGE C., Route 3, Plymouth, Wisconsin
- HITCHEN, BRIAN, Geological Survey, P.O. Box 671, Lusaka, Northern Rhodesia
- HJELMQVIST, SVEN, Dept. of Mineralogy, University, Lund, Sweden
- HOENES, DIETER, Geologisch-Mineralogisches Institut der Technischen Hochschule, Kaiserstrasse 12, Karlsruhe, Germany
- HOGARTH, DONALD D., Geological Survey of Canada, National Museum Building, Ottawa, Ontario, Canada
- HOLDEN, EARL, Box 245, Fairbanks, Alaska
- \*HOLDEN, GUERDON S., Plain Dealer Building, Cleveland, Ohio
- HOLLAR, ALBERT C., 3514 Taylor St. N.E., Minneapolis 18, Minnesota
- \*HOLMES, ARTHUR, University of Edinburgh, Grant Institute of Geology, King's Buildings, West Mains Rd., Edinburgh 9, Scotland
- HOLMES, M. A. (MRS.) 1 West 72 St., New York 23, N. Y.
- \*HOLMES, RALPH J., Dept. of Geology, Columbia University, New York 27, New York
- \*HOLSER, WILLIAM T., Institute of Geophysics, University of California, Los Angeles 24, California
- HONEA, RUSSELL M., 12 Geological Museum, Cambridge 38, Mass.
- HOOKER, MARJORIE, 2018 Luzerne Ave., Silver Spring, Maryland
- HOOVER, DONALD B., 2467 Stratford Rd., Cleveland Heights 18, Ohio
- HOPPIN, RICHARD A., Dept. of Geology, State University of Iowa, Iowa City, Iowa
- HOREN, ARTHUR, 6565 Rosemoor St., Pittsburgh 17, Pennsylvania
- HORN, AUBREY E., Care of F. H. Pigeon, P.O. Box 42, Jos, Plateau Province, Northern Nigeria
- HORNE, J. N. T., Geological Survey & Museum, Exhibition Rd., London S.W.7, England
- HOWELLS, LEWIS W., Box 661 Dante, Virginia
- HOWIE, R. A., Dept. of Geology, The University, Manchester 13, England
- \*HOWLAND, ARTHUR L., Dept. of Geology, Northwestern University, Evanston, Illinois
- Hsu, K. JINGHWA, Shell Development Co., 3737 Bellaire Blvd., Houston 25, Texas
- HUBBARD, BELA, 1405 E. Kleindale Rd., Tucson, Arizona
- HUGHES, WARREN E., Rt. 1, Box 8A, Hughson, California
- HUGHSON, MICHAEL R., 215 Patricia Ave., Ottawa 3, Ontario, Canada
- HUMMEL, FLOYD A., Dept. of Mineral Technology, Pennsylvania State University, University Park, Pennsylvania

- \*HUNT, WALTER F., Mineralogical Laboratory, University of Michigan, Ann Arbor, Michigan
- \*HURLBUT, CORNELIUS S., JR., Dept. of Mineralogy, Harvard University, Cambridge, 38, Massachusetts
- HURLEY DONAL, 16 Lewis St., Little Falls, New York
- HURST, THOMAS L., Research Dept., Monsanto Chemical Co., Columbia, Tennessee
- HURST, VERNON J., Dept. of Mines and Geology, 425 State Capitol, Atlanta, Georgia
- HUTCHINSON, R. W., 25 Adelaide St., W., Toronto, Ontario, Canada
- \*HUTTON, C. OSBORNE, Dept. of Mineral Sciences, Stanford University, Stanford, California
- ICHIMURA, TAKESHI, Institute of Earth Science, Faculty of Education, Yamagata University, Muikamachi, Yamagata-shi, Japan
- IMAI, HIKEKI, Tokyo University, Koman-Gakuka, Bunkyo-ku, Tokyo, Japan
- \*INGERSON, EARL, U. S. Geological Survey, Washington 25, D. C.
- INGRAM, W. F., P. O. Box 23, 433 West Taylor St., Griffin, Georgia
- \*INSLEY, HERBERT, 5219 Farrington Rd., Washington 16, D. C.
- INSLEY, ROBERT H., Champaion Spark Plug Co., 8525 Butler Ave., Detroit 11, Michigan
- INUZUKA, HIDEO, Matsuda Research Laboratory, Tokyo Shibaura Electric Co., Kawasaki, Japan
- IRWIN, WILLIAM H., 1075 Jasmine St., Denver 20, Colorado
- ISAACS, THELMA, 290 New Scotland Ave., Albany 8, New York
- ISHIBASHI, MASAO, Rigakubu-Chishitsu, Hokkaido Daigaku, Sapporo, Japan
- ISHIKAWA, HIDEO, Tokyo Kyoiku Daigaku, Chishitsugaku Kyoshitsu, Otsuku Kubomachi, Bunkyo-ku, Tokyo, Japan
- ISHIOKA, KOKICHI, Nagoya University at Mizuho, Nagoya, Japan
- ISOTOFF, A., Dept. of Geology, Idaho State College, Pocatello, Idaho
- JACKSON, KERN C., Dept. of Geology, University of Arkansas, Fayetteville, Arkansas
- JACKSON, M. L., Room 104B, Soils Building, University of Wisconsin, Madison 6, Wisconsin
- \*JACOBS, ELBRIDGE, 146 Williams St., Burlington, Vermont
- JACOBS, LOUIS J., The Obemayer Co., 2563 W. 18th St., Chicago 8, Illinois
- JAFFE, ARTHUR, 3212 W. Dauphin St., Philadelphia 32, Pennsylvania
- \*JAFFE, HOWARD W., 2910 Jennings Rd., Kensington, Maryland
- JAGO, JOHN B., 711 Kohl Building, San Francisco 4, California
- \*JAHNS, RICHARD H., Division of Geological Sciences, California Institute of Technology, Pasadena 4, California
- JAMES, C. C., Burford, Alexandra Place, Penzance, Cornwall, England
- \*JAMES, HAROLD L., U. S. Geological Survey, 4 Homewood Place, Menlo Park, California
- JARRELL, OSCAR W., 10 Nichols Rd., Needham 92, Massachusetts
- \*JEFFRIES, CHARLES D., Dept. of Agronomy, Agricultural Building, State College, Pennsylvania
- JENNESS, STUART E., 823 Archibald St., Ottawa, Ontario, Canada
- JENNI, CLARENCE M., 3129 Chadwick Drive, Los Angeles 32, California
- JENSEN, DAVID E., 199 East Brook Rd., Pittsford, New York
- JIZBA, ZDENEK V., 141-A South Milton Ave., Whittier, California
- JOHNE, C. T., Kragerö, Norway
- JOHNS, WILLIAM D., Dept. of Geology, Washington University, St. Louis, Missouri
- JOHNSON, CLAYTON H., 102 Swallow Hall, Columbia, Missouri
- JOHNSON, DONALD H., 43 Prospector Rd., Golden, Colorado

- \*JOHNSON, J. HARLAN, Colorado School of Mines, Golden, Colorado  
JOHNSON, RICHARD, 9 Wicker St., Ticonderoga, New York  
JOHNSON, ROBERT F., U.S.O.M., APO 676, New York, N. Y.  
JOHNSON, SARAH C., 1424 Somerset Place, N.W., Apt. 1, Washington, D. C.  
JOHNSON, W. M., RFD 6, Knoxville, Tennessee  
\*JOHNSTON, WILLIAM D., JR., U. S. Geological Survey, Washington 25, D. C.  
\*JOHNSTON, R. A. A., 112 Old Forest Hill Rd., Toronto 10, Ontario, Canada  
\*JOLLIFFE, ALFRED W., Dept. of Geology, Queens University, Kingston, Ontario, Canada  
JONAS, EDWARD C., Dept. of Geology, University of Texas, Austin 12, Texas  
JONES, FRANCIS T., 244 Trinity Ave., Berkeley 8, California  
JOSEPH, ED, Norwood, Colorado  
  
KADEY, FREDERIC L., JR., 1825 Quimby Lane, Westfield, New Jersey  
KAIMAN, S., 552 Booth St., Ottawa, Canada  
KAKITANI, MR., c/o Osaka Shiritsu Kaigaku, Rikogakubu Chigakukyoshitsu, Minami-Ogi-Cho, Kita ku, Osaka, Japan  
KALEY, MARY ELIZABETH, California Research Corporation, Box 543, Whittier, California  
KARRER, SEBASTIAN, Milwaukee Gas Specialty Co., P.O. Box 461, Milwaukee 1, Wisconsin  
KASEY, J. BRYANT, Box 968, Bakersfield, California  
KATSUI, Y., Chishitsu-Kobutsu Gaku-Kenkyushitsu, Rigakubu, Hokkaido University, Sapporo City, Japan  
KATZ, GERALD, Chemical Physics Branch, Signal Corps Eng. Labs., Fort Monmouth, New Jersey  
KAUFFMAN, A. J., JR., 1432 Dixon St., Corvallis, Oregon  
KEDESZY, H. H., 18 Laurelwood Drive, Little Silver, New Jersey  
KEENE, ARTHUR G., 124 South Parish Place, Burbank, California  
\*KEITH, MACKENZIE L., 204 Mineral Science Building, Pennsylvania State University, University Park, Pennsylvania  
\*KELLER, WALTER D., 208 Swallow Hall, Columbia, Missouri  
\*KELLEY, WALTER P., 120 Hilgard Hall, University of California, Berkeley 4, California  
KEMP, C. E., Saulte Ste. Marie Branch, Michigan College of Mining & Technology, Saulte Ste. Marie, Michigan  
\*KENNEDY, GEORGE C., Institute of Geophysics, University of California, Los Angeles, California  
\*KENNEDY, WILLIAM Q., Dept. of Geology, The University, Leeds 2, England  
\*KERR, PAUL F., Dept. of Geology, Columbia University, New York 27, New York  
KESSLER, CHARLES N., 317 South Holt Ave., Los Angeles 48, California  
KIDWELL, ALBERT L., Carter Oil Co., P.O. Box 801, Tulsa, Oklahoma  
KIENREICH, JOS. A., Sackstrasse 6, Graz, Austria  
KIETZMAN, WALTER L., Chile Exploration Co., Chuquicamata, Chile (via Antofagasta)  
KING, ELBERT A., JR., 2712 East 22 St., Austin, Texas  
KINGSBURY A. W. G., Dept. of Mineralogy, University Museum, Oxford, England  
KISSILEFF, M. Z., 329 Crest Park Rd., Philadelphia 19, Pennsylvania  
KLEEMAN, A. W., Mawson Laboratories, University of Adelaide, Adelaide, S. Australia  
KLEIN, GILBERT E., 402 Wesley Rd., Knoxville 19, Tennessee  
KLEINKOPF, M. DEAN, Standard Oil Co. of California, Box 278, Oildale, California  
KLINKENBERG, ROSEMARY, 884 Thomas Ave., St. Paul 4, Minnesota  
KNABE, WILLIAM C., 3853 North Gratz St., Philadelphia 40, Pennsylvania  
KNAPP, GUENEVER P., Geology Dept., Guyot Hall, Princeton University, Princeton, New Jersey  
KNIZEK, I. O., Apartado 2739, Mexico DF, Mexico



- \*KNOPF, ADOLPH, Dept. of Geology, Stanford University, Stanford, California  
KNUTSON, CARROLL F., c/o Continental Oil Co., 1137 Wilshire Blvd., Los Angeles 17, California  
KOARK, H. J., Geologiska Institut, Uppsala, Sweden  
\*KOSCHMANN, A. H., U. S. Geological Survey, Bldg. 25, Denver Federal Center, Denver Colorado  
KOBEL, CHARLES J., 9456 Grinnell St., Detroit 13, Michigan  
KOHN, JACK A. Chemical-Physics Branch, Signal Corps Engineering Laboratory, Fort Monmouth (Hexagon), New Jersey  
KONTA, JIRI, Institute of Petrology, Albertov 6, Praha II, Czechoslovakia  
KOUCKY, FRANK, 5112 Ingleside, Chicago 15, Illinois  
\*KRAUS, EDWARD H., Mineralogical Laboratory, University of Michigan, Ann Arbor. Michigan  
\*KRISHNAN, MAHARAJAPURAM S., 27 Chowringhee, Calcutta, India  
KRPATA, FRANCIS, 61 Harris Ave., Hewlett, New York  
\*KRYNINE, PAUL D., Dept. of Mineralogy, Pennsylvania State University, University Park, Pennsylvania  
KRYSIK, STANLEY, 57 Keele St., Toronto 9, Ontario, Canada  
KULLERUD, GUNNAR, 5520 Carolina Place, N.W., Washington 16, D. C.  
\*KULP, J. LAURENCE, Dept. of Geology, Columbia University, New York 27, N. Y.  
KUNO, HISASHI, Geological Institute, Faculty of Science, Tokyo University, Tokyo, Japan  
KUNZE, GEORGE W., Agronomy Dept., Texas A. & M. College, College Station, Texas  
KURBATOV, JOANN D., Dept. of Physics, Ohio State University, Columbus, Ohio  
KURDEKA, PETER L., 26 George St., Thomaston, Connecticut  
KUTINA, JAN, Mineralogical Institute, Charles University, Albertov 6, Praha II, Czechoslovakia
- LACY, WILLARD C., La Oroya, Peru  
LADOO, RAYMOND B., 42 Huntington Rd., Newton 58, Massachusetts  
LAKIN, HUBERT W., 860 Dudley St., Denver 14, Colorado  
\*LANDES, KENNETH K., Geology Dept., University of Michigan, Ann Arbor, Michigan  
LANG, ANDREW J., JR., Dept. of Geology, University of Wichita, Wichita 6, Kansas  
LANGFORD, LAWRENCE, Asan Point Community Center, Station S, Asan, Guam  
LAPKOWSKY, W. W., 205 Stanley Ave., Hamilton, Ontario, Canada  
LARKIN, ROBERT G., 649 Bienvenida Ave., Pacific Palisades, California  
\*LARSEN, ESPER S., JR., Apartment 502-H, 3920 Connecticut Ave., N.W., Washington 8, D. C.  
\*LARSEN ESPER S., 3d, 7508 Radnor Rd., Bethesda 14, Maryland  
LAVES, FRITZ, Mineralogical Institute, E. T. H., Sonneggstrasse 5, Zürich 6, Switzerland  
LAWRENCE, EDMOND F., Box 9044, University Station, Reno, Nevada  
\*LEE, HARLEY C., Basic Refractories, Inc., 845 Hanna Bldg., Cleveland 15, Ohio  
LEIGHTON, MORRIS W., Carter Oil Co., Research Laboratory, Box 801, Tulsa, Oklahoma  
\*LEMMON, DWIGHT M., U. S. Geological Survey, Washington 25, D. C.  
LEONARD, BENJAMIN F., U. S. Geological Survey, Denver Federal Center, Denver, Colorado  
LEPP, HENRY, 925 East Seventh St., Duluth 5, Minnesota  
LETORT, YVES, 3, Avenue Paul Doumer, Paris 16, France  
\*LEVIN, S. BENEDICT, 50 Crest Drive, Little Silver, New Jersey  
LEVINE, HARRY, 552 Nicholson St., NE., Washington, D. C.  
LEVINSON, ALFRED A., Dept. of Mineralogy, Ohio State University, Columbus, Ohio  
LEWIS, EARL E., Box 303, Klickitat, Washington



- LIDDICOAT, RICHARD T., JR., 541 South Alexandria Ave., Los Angeles 5, California
- LIGHT, MITCHELL A., California Research Corp., Box 446, La Habra, California
- LINCOLN, EARLE R., Holberg, British Columbia, Canada
- \*LINDBERG, MARIE LOUISE (MRS.), U. S. Geological Survey, Washington 25, D. C.
- LINDHE, HENRY E., P.O. Box 130, Austin 2, Texas
- LINDQUIST, BENGT, Mineralogisk-Geologisk Institut, F. D. Konsistoriehuset, Uppsala, Sweden
- LINVILLE, P. E., 6258 Mulholland Highway, Hollywood 28, California
- \*LIPSCOMB, WILLIAM N., School of Chemistry, University of Minnesota, Minneapolis 14, Minnesota
- LIVINGSTON, JOHN J., 731 Eighth St., Marysville, California
- LOEWENSTEIN, WALTER, Rua Bento Pereira, 326 Casa 1, Sao Paulo, Brazil
- LOMBARDI, ORESTE W., 2847 Reposa Lane, Altadena, California
- \*LONSDALE, KATHLEEN, University College, Gower St., London W.C.1, England
- \*LONSDALE, JOHN T., Bureau of Economic Geology, University of Texas, Austin 12, Texas
- \*LOPEZ, VICTOR M., Apartado 3585, Caracas, Venezuela
- \*LOUDERBACK, GEORGE D., University of California, Berkeley 4, California
- LOUPEKINE, I. L., Dept. of Geology, The University, Bristol 8, England
- \*LOVERING, THOMAS, S., 8001 West 17th Ave., Lakewood, Colorado
- LOWE, KURT E., 49-01 Francis Lewis Blvd., Bayside 64, New York
- LOWE, W. ROSS, Sudbury Assay Office, 256 Oak St., Sudbury, Ontario, Canada
- LUCAS, ELMER L., 219 West Eufala St., Norman, Oklahoma
- LUEDEMANN, LOIS WEISER, 431 West Fairmount Ave., State College, Pennsylvania
- \*LUKESH, JOSEPH S., 516 Riverside Ave., Scotia, New York
- LUND, JAMES R. (US 55493452), 519th Engineer Co. Depot, APO 331, San Francisco, California
- LYDEN, EDWARD F. X., 86 Spring Valley Rd., Park Ridge, New Jersey
- LYON, CRAIG A., 3300 Kingman Blvd., Des Moines, Iowa
- LYONS, JOHN B., 7 Pleasant St., West Lebanon, New Hampshire
- LYONS, S. C., Georgia Kaolin Co., Dry Branch, Georgia
- MACDONALD, GORDON A., U. S. Geological Survey, Hawaiian Volcano Observatory, Hawaii Volcano Observatory, Hawaii National Park, T. H.
- \*MACDONALD, GORDON J. P., Dept. of Geology, Massachusetts Institute of Technology, Cambridge 39, Massachusetts
- MACKAY, RALPH E., 2923 East Harrison St., Seattle 2, Washington
- MACKENZIE, DAVID B., American Overseas Petroleum, Ltd., 380 Madison Ave., New York 17, New York
- MACKENZIE, G. S., Dept. of Geology, University of New Brunswick, Fredericton, N. B., Canada
- MACKENZIE, ROBERT C., Macaulay Institute of Soil Research, Craigiebuckler, Aberdeen, Scotland
- MACKENZIE, WILLIAM SCOTT, Geophysical Laboratory, 2801 Upton St., N. W., Washington 8, D. C.
- MACKERACHER, D. W., Apt. A14, 1546 Kingston Rd., Toronto 13, Ontario, Canada
- MACLACHLAN, J. C., U. S. Geological Survey, Denver Federal Center, Denver, Colorado
- MACLAREN, ALEXANDER S., 138 Second Ave., Apt. 8, Ottawa, Ontario, Canada
- MAJOR, MILLARD H., 817 Wilson Tower, Corpus Christi, Texas
- MANDARINO, JOSEPH A., Mineralogical Laboratory, University of Michigan, Ann Arbor, Michigan
- MARCIN, EDWARD J., 1917 Meadowbrook Rd., North Merrick, Long Island, New York

- MARINOS, GEORGE, Aminandrou 31 Koukaki, Athens, Greece
- MARKMAN, HARVEY C., The Colorado Museum of Natural History, City Park, Denver 6, Colorado
- MARTIN, E. B., Westbury, Long Island, New York
- MASON, VINCENT I., 154 Beaumont Ave., Newtonville 60, Massachusetts
- \*MARSHALL, C. EDMUND, Dept. of Soils, College of Agriculture, University of Missouri, Columbia, Missouri
- \*MARTENS, JAMES H. C., Bureau of Mineral Research, Rutgers University, New Brunswick, New Jersey
- \*MASON, BRIAN H., Dept. of Geology, American Museum of Natural History, New York 24, New York
- MATHER, MRS. BRYANT, P.O. Drawer 2131, Jackson, Mississippi
- MATHEWSON, HARRIS D., 14368 Bora Drive, La Mirada, California
- MATSUMOTO, HATAO, Geological Institute, Kumamoto University, Kumamoto, Kyushu, Japan
- MATTHEWS, FRED W., Canadian Industries, Ltd., Central Research Laboratory, McMasterville, Quebec, Canada
- MATTOX, RICHARD B., Dept. of Geology, Texas Technological College, Lubbock, Texas
- MATZKO, JOHN J., U. S. Geological Survey, 4 Homewood Place, Menlo Park, California
- \*MAUGUIN, CHARLES, 14 Ave. Hoche, Thiais, Seine, France
- MAURICE, CHALFS S., 215 Cameron Ave., Chapel Hill, North Carolina
- MAWBY, M. A., 95 Collins St., Melbourne, Victoria, Australia
- \*MAYNARD, JAMES E., Dept. of Mineralogy, Syracuse University, Syracuse, New York
- MCALPIN, ARCHIE, Notre Dame University, Notre Dame, Indiana
- MCANDREW, JOHN CSIRO, Geology Dept., University of Melbourne, Carlton N 3, Victoria, Australia
- MCATEE, JAMES L., JR., Baroid Sales Division, National Lead Co., Box 1675, Houston 1, Texas
- McBURNAY, T. C., Smith-Emery Co., 781 East Washington Blvd., Los Angeles 21, California
- McCAIG, DONALD W., 217 Cagua Drive NE, Albuquerque, New Mexico
- McCALL, KEITH E., JR., 311 Loma Ave., Syracuse, New York
- \*McCAUGHEY, WILLIAM J., Lord Hall, Ohio State University, Columbus 10, Ohio
- \*McCONNELL, DUNCAN, Dept. of Mineralogy, Lord Hall, Ohio State University, Columbus 10, Ohio
- McCREERY, G. L., Technical Director, Ohio Brass Co., Box 11, Barberton, Ohio
- McCREERY, R. A., Apartado 51, Instituto, Turrialba, Costa Rica, C. A.
- McGARRY, FRED M., 447 W. 24th Ave., Spokane, Washington
- McGIRK, LON S., JR., Mackay School of Mines, Reno, Nevada
- McILHENNY, GEORGE N., Box 248, Lake, Mississippi
- McINTOSH, ALEX O., Purvis Club, McMasterville, Quebec, Canada
- \*McKINSTRY, HUGH E., Vanserg Laboratories, Cambridge 38, Massachusetts
- \*McLACHLAN, DAN, JR., Stanford Research Institute, Stanford, California
- \*McLAUGHLIN, DONALD H., 100 Bush St., 26th floor, San Francisco 4, California
- McLAUGHLIN, R. J. W., Dept. of Mineralogy, Downing St., Cambridge, England
- McLELLAN, ROY D., 24 Lillian Terrace, Woodbridge, New Jersey
- \*McMURDIE, HOWARD F., National Bureau of Standards, Washington, D. C.
- McNEIR, THOMAS S., 9 Governors Rd., Bronxville 8, New York
- \*McVAY, THOMAS N., 10 Oakwood Court, Tuscaloosa, Alabama
- McVEAGH, WILLIAM J., 527½ Gettysburg St., Pittsburgh 6, Pennsylvania
- MEEK, WARD BENJAMIN, 6400 West 6th Ave., Denver 14, Colorado

- \*MEEN, V. BEN, Royal Ontario Museum of Geology and Mineralogy, 100 Queen's Park, Toronto 5, Ontario, Canada
- \*MEIXNER, HEINZ, Bergdirektion Hüttenberg, Knappenberg, Kärnten, Austria
- MELLIS, OTTO, Stocholms Hogskolas Mineralogiska Inst., Kungstengatan 45, Stockholm, Sweden
- MENCHER, ELY, Dept. of Geology and Geophysics, Massachusetts Institute of Technology, Cambridge 39, Massachusetts
- MERGNER, JOHN L., 5519 Nevada Ave., N.W., Washington, D. C.
- \*MERRITT, CLIFFORD A., University of Oklahoma, Norman, Oklahoma
- MERRITT, PHILLIP L., 450 Clinton Ave., Brooklyn 5, New York
- \*MERTIE, JOHN B., JR., U. S. Geological Survey Building, Agricultural Research Center, Beltsville, Maryland
- \*MERWIN, HERBERT E., Geophysical Laboratory, 2801 Upton St., N.W., Washington 8, D. C.
- \*MEYER, CHARLES, 104 Bacon Hall, University of California, Berkeley 4, California
- MEYERS, T. R., Dept. of Geology, University of New Hampshire, Durham, New Hampshire
- MEYROWITZ, ROBERT, 12905 Bluhill Rd., Silver Spring, Maryland
- \*MICHENER, CHARLES E., International Nickel Company of Canada, Ltd., Copper Cliff, Ontario, Canada
- MIDGLEY, H. G., Building Research Station, Garston, Watford, Herts, England
- \*MIELENZ, RICHARD C., Bureau of Reclamation, Denver Federal Center, Denver, Colorado
- MIKAMI, HARRY M., E. J. Lavino & Co., Box 29, Norristown, Pennsylvania
- MILLER, CLARENCE E., Dept. of Geology & Geography, University of Rhode Island, Kingston, Rhode Island
- MILLER, F. STUART, 49 Mendota Ave., Rye, New York
- MILLER, ROSWELL, 3d, 518 Felt Bldg., Salt Lake City, Utah
- \*MILLER, WILLIAM J., 520 Baughman Ave., Claremont, California
- MILLMAN, A. P., Dept. of Mining Geology, Royal School of Mines, London S.W.7, England
- MILNE, IVAN H., 832 12th St., Oakmont, Pennsylvania
- \*MILTON, CHARLES, U. S. Geological Survey, Washington 25, D. C.
- MINATO, HIDEO, c/o Tokyo Daigaku Kyoyogakubu, Komaba, Meguro-ku, Tokyo, Japan
- \*MISER, HUGH D., 4025 South Dakota Ave., N.E., Washington 18, D. C.
- MITCHELL, RICHARD, Dept. of Geology, University of Virginia, Charlottesville, Virginia
- MITSUDA, T., Geological Institute, Faculty of Science, Yamaguchi University, Itoyone, Yamaguchi City, Japan
- MITSUI, KYO, Kenai-Jutaku Dai 2-go, Hatogaoka, Hitachi City, Ibaragi Pref., Japan
- MODDLE, DOUGLAS A., Dept. of Mines, Parliament Bldgs., Queens Park, Toronto, Canada
- \*MONEYMAKER, BERLEN C., c/o T.V.A. Geologic Division, 620 Union Bldg., Knoxville, Tennessee
- \*MONTGOMERY, ARTHUR, 277 Park Ave., New York City
- \*MOORE, ELWOOD S., University of Toronto, Toronto 5, Canada
- MORGAN, LEONARD A., 217 South Atlantic Avenue, Haddonfield, New Jersey
- MORGRIDGE, DEAN L., Humble Oil and Refining Co., 610 High St., Eugene, Ohio
- MORIKAWA, PROF., Shigadaigaku, Gakugei-Gakubu, Zeze, Otsu City, Japan
- MORIMOTO, RYOHEI, Earthquake Research Institute, Tokyo University, Tokyo, Japan
- MORLEY, RUSSELL A., 399 North 18 St., Salem, Oregon
- \*MORRIS, FREDERICK K., Arctic Desert Tropic Info Center, R.S.I., Maxwell AFS, Alabama
- MORRISON, BERT C., American Smelting & Refining Co., Patagonia, Arizona
- MORTOLA, DRA. EDELMIRA, Junin 1357, Piso 3, A, Buenos Aires, Argentina

- MOYD, LOUIS, 136 Park Ave., Yonkers 3, New York
- \*MROSE, MARY E., U. S. Geological Survey, Washington 25, D. C.
- MULLI, HANS, Mineralogical Institute, E.T.H., Sonneggstrasse 5, Zürich 6, Switzerland
- MULLNER, HENRY C., 141 Ackerman Ave., Clifton, New Jersey
- MUMPTON, FRED A., 505 Tulip Road, State College, Pennsylvania
- MURAOKA, MAKOTO, Matsuo Mining Co., Ltd., New Marunouchi Bldg., Marunouchi Chiyoda-ku, Tokyo, Japan
- \*MURATA, K. J., U. S. Geological Survey, Washington 25, D. C.
- \*MURDOCH, JOSEPH, University of California at Los Angeles, Los Angeles 24, California
- \*MUROWCHICK, BERNARD L., 637 Sagamore St., Lakeland, Florida
- MURRAY, ALBERT N., Dept. of Geology, University of Tulsa, Tulsa 4, Oklahoma
- MURRAY, HAYDN H., Dept. of Geology, Indiana University, Bloomington, Indiana
- MURPHY, RICHARD, Newmont Mining Corp. of Canada, 25 King St. West, Toronto, Canada
- MUTCH, A. D., Hardy Mine, Levack, Ontario, Canada
- MYERS, ALFRED T., U. S. Geological Survey, Denver Federal Center, Denver, Colorado
- MYERS, HENRY C., 2308 National Ave., Palm City, California
- MYERS, PAUL E., Box 35, King Ferry, New York
- \*NAGELSCHMIDT, GUNTER, Birchtrees, Ninelands, Hathersage near Sheffield, England
- NAIDU, P. R. J., Dept. of Geology, University of Madras, Madras 25, S. India
- NAIR, K. V. Krishnan, Division of Mineral Survey & Research, University of Travancore Trivandrum, India
- NALLE, PETER B., 4600 Indian Hill Rd., Riverside, California
- NASON, HOWARD K., 1700 South Second St., St. Louis 4, Missouri
- NAVIAS, ROBERT A., 204 East Eighth St., Leadville, Colorado
- NEIMAN, ROBERT, 2014 Lakeside Drive, Louisville 5, Kentucky
- \*NEL, HANS JURIE, Central Metallurgical Laboratory, Anglo American Corp. of South Africa, Ltd., Box 11181, Johannesburg, South Africa
- NELSON, HARRY E., 1018 East Norman Ave., Las Vegas, Nevada
- \*NEUERBURG, GEORGE J., U. S. Geological Survey, Denver Federal Center, Bldg. 25, Denver, Colorado
- \*NEWHOUSE, WALTER H., Dept. of Geology, University of Chicago, Chicago 37, Illinois
- NIES, NELSON P., 1495 Coolidge Ave., Pasadena, California
- NIESET, CARL F., St. Joseph College, Collegeville, Indiana
- NIGGLI, E., Mineralogisch-Petrographisches Institut der Universität Bern, Sahlistrasse 6, Bern, Switzerland
- NISSON, WILLIAM H., 815 D St., Petaluma, California
- \*NOBLE, JAMES A., California Institute of Technology, Pasadena 4, California
- \*NOLAN, THOMAS B., U. S. Geological Survey, Washington 25, D. C.
- NORIN, ROLF, Hoganas Billeholms Aktiebolag Biblioteket, Hoganas, Sweden
- NORMART, JOSEPH J., 438 South 3rd Ave., Tucson 1, Arizona
- NORTON, JAMES J., U. S. Geological Survey, Denver Federal Center, Denver, Colorado
- \*NOWACKI, WERNER, Mineralogisch-Petrographisches Institut, Universität Bern, Sahlistrasse 6, Bern, Switzerland
- NOZAWA, KAZAHISA, 111 Shiba-Shirokane-Sankocho, Minato-ku, Tokyo, Japan
- \*NUFFIELD, EDWARD W., Dept. of Geological Sciences, University of Toronto, Toronto 5, Canada
- NYGREEN, PAUL W., Box 1660, Midland, Texas
- OBERHOLZER, WALTER, Rua Castilho 209-2 E, Lisbon, Portugal



- O'DANIEL, H., Mineralogisches Institut der Universität, Senckenberg Anlage 30, Frankfurt, Germany
- ODOM, W. R., Star Rt. A, Box 32-C, Austin, Texas
- OKE, WILLIAM C., 9115 South Harvard Boulevard, Los Angeles 47, California
- O'LEARY, W. J., 24, Seminole, Claymont, Delaware
- OLSON, J. C., U. S. Geological Survey, Washington 25, D. C.
- OMORI, KEI-ICHI, Institute of Mineralogy, Petrology and Economic Geology, Tohoku University, Katahiracho, Sendai, Japan
- ÖNAY, TOGAN Ş., Gazi Bulvari 38, Sokak no. 15, Aydin, Turkey
- OOTSUBO, YOSHIO, 10-22, Nishikata-machi, Bunkyo-ku, Tokyo, Jaapn
- ORDWAY, FRED, 5205 Elsmere Ave., Bethesda 14, Maryland
- ORTEGA, FRANCISCO BARROSO, Lab. No. 2 de Mineralogia, Universidad de la Habana, Habana, Cuba
- OSBERG, PHILIP H., 122 Mayflower Hill Drive, Waterville, Maine
- OSBORN, DOROTHY S., 2812 East 5th Ave., Spokane, Washington
- \*OSBORN, ELBURT F., Dept. of Earth Sciences, School of Mineral Industries, Pennsylvania State University, University Park, Pennsylvania
- \*ORCEL, JEAN, Museum d'Histoire Naturelle, 61 Rue de Buffon, Paris V, France
- OSTHAUS, B. B., 1108 Pacific Ave., Brackenridge, Pennsylvania
- OVER, EDWIN, Box 596, Woodland Park, Colorado
- OVERMAN, WILLIAM, Rt. 1, Chapel Hill, North Carolina
- \*PABST, ADOLF, University of California, Berkeley 4, California
- PAINTER, HOWARD S., 936 Fullerton Ave., Williamsport, Pennsylvania
- \*PARDILLO, FRANCISCO, University of Barcelona, Barcelona, Spain
- \*PAGE, LINCOLN R., 4101 Saul Rd., Chevy Chase View, Kensington, Maryland
- \*PARK, CHARLES F., JR., Dept. of Geology, Stanford University, Stanford, California
- PARKER, JOHN G., 2L Laurel Hill Rd., Greenbelt, Maryland
- \*PARKER, ROBERT L., Technische Hochschule, Sonneggstr. 5, Zürich 6, Switzerland
- PARNAU, JOHN L., 1342 North Lincoln St., Stockton, California
- \*PARRISH, WILLIAM, Philips Laboratories, Inc., Irvington-on-Hudson, New York
- \*PARSONS, ARTHUR L., Royal Ontario Museum of Mineralogy, 100 Queen's Park, Toronto 5, Ontario, Canada
- \*PARSONS, WILLARD H., Dept. of Geology, Wayne University, Detroit 1, Michigan
- PASK, JOSEPH A., Division of Mineral Technology, University of California, Berkeley 4, California
- \*PATERSON, A. L., Institute for Cancer Research, Fox Chase, Philadelphia 11, Pennsylvania
- PAULING, LINUS, California Institute of Technology, Pasadena, California
- PEACOR, DONALD, 4 Hillside Ave., Stoneham 80, Massachusetts
- PEARCE, D. W., 1814 Hunt, Richland, Washington
- PEARL, RICHARD M., Colorado College, Colorado Springs, Colorado
- PEARSON, ROBERT C., U. S. Geological Survey, Bldg. 25, Denver Federal Center, Denver, Colorado
- \*PECORA, WILLIAM T., U. S. Geological Survey, Washington 25, D. C.
- \*PEGAU, ARTHUR A., 1808 Winston Ave., Charlottesville, Virginia
- PELTO, CHESTER R., MRD Laboratory, Corps of Engineers, 18th & Howard Sts., Omaha 2, Nebraska
- PENDLETON, NORMAN H., 111 Pryce St., Santa Cruz, California
- PENDRAY, GUENEVER P., 264 Hawthorne Ave., Princeton, New Jersey
- PENG, C. J., 724 West Dayton St., Madison, Wisconsin



- \*PEOPLES, JOE WEBB, U. S. Geological Survey, American Embassy, APO 928, San Francisco, California
- \*PEPINSKY, RAYMOND, Research Professor of Physics, Pennsylvania State University, University Park, Pennsylvania
- PERLOFF, LOUIS, 1831 Nissen Building, Winston-Salem, North Carolina
- PERMINGEAT, FRANÇOIS, Service Géologique, Rabat, Morocco
- PERRY, ELWYN L., Dept. of Geology, Williams College, Williamstown, Massachusetts
- PERUSEK, CYRIL J., 2405 W. Cuthbert, Midland, Texas
- PETERSON, NELS P., Box 1211, Globe, Arizona
- PAHAIR, GEORGE, U. S. Geological Survey, Washington 25, D. C.
- \*PIEMISTER, T. C., Dept. of Geology and Mineralogy, University of Aberdeen, Marischal College, Aberdeen, Scotland
- PHILBROOK, P. M. (Mrs.), P.O. Box 184, Sacramento 1, California
- PHILLIPS, WILLIAM R., 2756 Adams St., Salt Lake City, Utah
- PIERSON, CHARLES T., U. S. Geological Survey, P.O. Box 360, Grand Junction, Colorado
- PINSON, WILLIAM H., JR., 52 Hillside Rd., Watertown, Massachusetts
- PLATT, PHILIP L., 74 Beacon Hill Rd., Port Washington, New York
- POCOVI, ANTONIO, Rivadavia 3567, Santa Fe, Argentina
- POHOST, GERALD, 5373 Chillum Place N.E., Washington, D. C.
- POIRE, INNA V., U. S. Geological Survey, Washington 25, D. C.
- \*POLDERVAART, ARIE, Dept. of Geology, Columbia University, New York 27, New York
- POLNAK, ROBERTO J., Mitre 828, Berazategui, Buenos Aires, Argentina
- POOLE, S. W., 1346 22 St. NW, Canton 9, Ohio
- PORTO, JUAN C., Suipacha 588, San Miguel de Tucuman, FCNGEM, Argentina
- \*POSTEL, A. WILLIAMS, 3703 Farragut St., Kensington, Maryland
- \*POUGH, FREDERICK H., 4680 Independence Ave., New York 71, New York
- PRATHER, ROBERT W., U. S. Navy Electronics Laboratory, San Diego 52, California
- PRENGRAVE, CYRIL, Gwynedd Valley, Pennsylvania
- PRICE, EUGENE W., 1705 South 69th, West Allis 14, Wisconsin
- PRICE, THOMAS, Price, North Dakota
- \*PRIDER, REX T., Dept. of Geology, University of Western Australia, Nedlands, Western Australia
- PRIEN, EDWIN L., 1101 Beacon St., Brookline 46, Massachusetts
- PUTNAM, BENJAMIN H., 606 Third St., Marietta, Ohio
- \*QUENSEL, PERCY D., Stockholms Högskola, Stockholm, Sweden
- \*QUINN, ALONZO W., Dept. of Geology, Brown University, Providence 12, Rhode Island
- \*RABBITT, JOHN C., U. S. Geological Survey, Washington 25, D. C.
- RAHKO, HANNES, Rt. 1, Box 1560, Clarkston, Washington
- RAISCH, WILLIAM, 86-56 Palo Alto St., Hollis, Long Island, New York
- \*RAMBERG, HANS, Dept. of Geology, University of Chicago, Chicago 37, Illinois
- \*RAMDOHR, PAUL, Mineralogisch-Petrographisches Institut der Universität, Hauptstrasse 47-51, Heidelberg, Germany
- RAMISCH, J. L., 419 Belton Rd., Silver Spring, Maryland
- \*RAMSDELL, LEWIS S., Mineralogical Laboratory, University of Michigan, Ann Arbor, Michigan
- \*RANKAMA, KALERVO, Institute of Geology, University of Helsinki, Snellmanink 5, Helsinki, Finland
- RATH, ROBERT, Königswortherstrasse 2 III, (20a) Hannover-W, Germany

- RATTÉ, JAMES C., U. S. Geological Survey, Mineral Deposits Branch, Denver Federal Center, Denver, Colorado
- REA, ROBERT F., Stupakoff Ceramic Mfg. Co., Latrobe, Pennsylvania
- REDDEN, JACK A., U. S. Geological Survey, Mineral Deposits Branch, Denver Federal Center, Denver, Colorado
- REED, FRANK J., 3118 Rittenhouse St., N.W., Washington, D. C.
- REEVES, AUGUSTUS, Box 1474, Salt Lake City, Utah
- REICHEN, LAURA E., U. S. Geological Survey, Washington 25, D. C.
- REITAN, PAUL H., Dept. of Geology, University of Chicago, Chicago 37, Illinois
- REMICK, JEROME H., 3d, 157 Ridge Rd., Grosse Pointe Farms 36, Michigan
- RENFRO, GEORGE E., 142-49 41 Ave., Flushing, New York
- REYES, JORGE MUNOX, Apartado 267, La Paz, Bolivia
- RICE, H. M., Chemistry Division, Science Service Bldg., Carling Ave., Ottawa, Canada
- RICHMOND, JAMES F., Division of Natural Sciences, Los Angeles State College, 855 N. Vermont Ave., Los Angeles, California
- \*RICHMOND, WALLACE E., JR., RD 2, Hillsboro, New Hampshire
- RICHTER, DONALD H., U. S. Geological Survey, Box 654, Camaguay, Cuba
- RICKER, R. W., 341 Vernon St., New Kensington, Pennsylvania
- \*RIDGE, JOHN D., Lemont, Centre County, Pennsylvania
- RIGSBY, GEORGE P., SIPRE, 1215 Washington Ave., Wilmetta, Illinois
- RILEY, L. B., U. S. Geological Survey, Denver Federal Center, Bldg. 25, Denver, Colorado
- RINN, HAROLD W., 2913 Braley Court, Midland, Michigan
- RISKA, DAPHNE DEE, 410 Cedar St., N. W., Apt. 25, Washington 12, D. C.
- RITCHIE, EVERETT J., Eagle-Picher Company, Research Laboratories, Joplin, Missouri
- ROIROIR, LUIS, Consejo Superior de Investigaciones Científicas, Instituto de Fíxica, "Alonso de Santa Cruz," Serrano 119, Madrid, Spain
- ROBBINS, CARL R., National Bureau of Standards, Div. 9.7, Washington 25, D.C.
- ROBERTS, FRED, JR., 1106 West Newmark, Monterey Park, California
- ROBERTS, ORAN M., Box 7712, University Station, Austin, Texas
- ROBERTS, W. L., 628 Harter Drive, Rapid City, South Dakota
- ROBERTSON, E. C., Acorn Building, Eastern Ave., and Newell St., Silver Spring, Maryland
- ROBERTSON, FORBES, Dept. of Geology, University of Washington, Seattle, Washington
- ROBERTSON, ROBERT H. S., 16 Kirklee Road, Glasgow W. 2, Scotland
- ROBIE, RICHARD A., Dept. of Geology, University of Chicago, Chicago 37, Illinois
- ROBINSON, G. D., General Geology Branch, U. S. Geological Survey, Denver Federal Center, Denver, Colorado
- ROBINSON, STEPHEN C., Geological Survey of Canada, Victoria Memorial Museum, Ottawa, Ontario, Canada
- RODE, K. P., University Department of Geology, M B College, Udepur, India
- RODEKOH, W. J., 25 Elgin St., Alhambra, California
- \*ROEDDER, EDWIN W., U. S. Geological Survey, Washington 25, D. C.
- \*ROGERS, AUSTIN F., 2423 Virginia St., Berkeley, California
- ROGERS, CLEAVES L., American Embassy, La Fragua 18, Mexico, D. F., Mexico
- ROLSHAUSEN, F. W., P.O. Box 2180, Houston 1, Texas
- ROONEY, JOHN T., Box 358, Buffalo, New York
- ROSE, ROBERT L., 1519 Milvia St., Berkeley 9, California
- ROSENBLUM, SAM, U. S. Geological Survey, Denver Federal Center, Denver, Colorado
- ROSENFELD, JOHN L., Wesleyan University, Dept. of Geology, Middleton, Connecticut
- \*ROSENHOLTZ, JOSEPH L., Rensselaer Polytechnic Institute, Troy, New York
- ROSENKRANS, R. R., 373 Albert, Shreveport, Louisiana

- ROSENZWEIG, ABRAHAM, Dept. of Geology, University of New Mexico, Albuquerque, New Mexico
- \*ROSS, CLARENCE S., U. S. Geological Survey, Washington 25, D. C.
- ROSS, VIRGINIA F. (MRS.), Dept. of Chemistry, Brown University, Providence, Rhode Island
- ROTH, ROBERT S., 8000 Newell Court, Silver Spring, Maryland
- \*ROWLAND, RICHARDS A., Shell Oil Company, P.O. Box 2099, Houston 1, Texas
- ROWLAND, ROBERT W., Natural History Museum, Balboa Park, San Diego 1, California
- ROWLEY, ELMER B., 214 Ridge St., Glens Falls, New York
- ROY, CHALMER J., Dept. of Geology, Iowa State College, Ames, Iowa
- \*ROY, RUSTUM, 308 Mineral Sciences Building, Pennsylvania State University, University Park, Pennsylvania
- ROYAL, H. F., Champion Spark Plug Co., 8525 Butler Ave., Detroit 11, Michigan
- ROYER, LOUIS, Laboratoire de Minéralogie, Université, Alger, Algeria
- RUBENSTEIN, SAMUEL, U. S. Geological Survey, Washington 25, D. C.
- RUGGLES, RICHARD D., 30 Carleton St., Revere 51, Massachusetts
- \*RUNNER, JOSEPH J., Geology Building, University of Iowa, Iowa City, Iowa
- \*RUSSELL, SIR ARTHUR, Swallowfield Park, Reading, Berkshire, England
- RUSSELL, ROBERT T., 2442 King St., Denver 11, Colorado
- RYAN, J. DONALD, Dept. of Geology, Lehigh University, Bethlehem, Pennsylvania
- SABINE, P. A., 249 Boston Manor Rd., Brentford, Middlesex, England
- SACHS, WALTER P., 254 Bloomfield Ave., Caldwell, New Jersey
- SAEGUSA, MORITSUNA, Taihei Kogyo KK, Akenobe Kogyo-sho, Minami-dani-mura, Yabu-gun, Hoygo-ken, Japan
- SAFFORD, ARTHUR T., JR., Silvermine Rd., New Canaan, Connecticut
- \*SAHAMA, THURE GEORG, Institute of Geology, Snellmanink 5, Helsinki, Finland
- SAHLIN, CARL INGEMAR, Willastigen 1, Kiruna, Sweden
- SAKURA, KIYOSHI, c/o Nihon Mining Co., Ltd., Minenosawa Kogyo-sho, Tatsuyama-mura, Iwate-gun, Shizuoka-ken, Japan
- SALAMON, M., Medical Laboratory, 100 Allenby Rd., Tel Aviv, Israel
- SAMEJIMA, TERUHIKO, Shizuoka Univ. Bunrigakubu, Chighitsugaku-Kyoshitsu, Ooiwa-cho, Shizuoka, Japan
- \*SAMPSON, EDWARD, Guyot Hall, Princeton University, Princeton, New Jersey
- SAMPTER, E. L., 49 West 45th St., New York 19, N. Y.
- SAND, L. B., Standard Oil Company (Indiana), Engineering Research Dept., 2400 New York Ave., Whiting, Indiana
- \*SANDELL, ERNEST B., School of Chemistry, University of Minnesota, Minneapolis 14, Minnesota
- SAN MARTÍN Y SÁENZ, RENÉ, Laboratorio de Mineralogia No. 1, Facultad de Ciencias, Universidad de la Habana, Habana, Cuba
- SARAKAUSKAS, RAYMOND V., National Carbon Research Laboratory, Box 6056, Cleveland, Ohio
- SAVOLAHITI, ANTTI O. M., Jaakarinkatu 10a 12, Helsinki, Finland
- SAYLOR, PAULINE A., 20144 East Arrow Highway, Covina, California
- \*SCHAIRER, JOHN F., Geophysical Laboratory, 2801 Upton St., N.W., Washington 8, D. C.
- \*SCHALLER, WALDEMAR T., U. S. Geological Survey, Washington 25, D. C.
- \*SCHMITT, HARRISON A., Cottage Sanatorium Rd., Silver City, New Mexico
- SCHAUB, STANLEY J., 105 Elm St., Westfield, New Jersey
- SCHEID, VERNON E., Mackay School of Mines, University of Nevada, Reno, Nevada
- SCHERRER, PETER, 4032 Egbert Ave., Cincinnati 20, Ohio

- SCHEUMANN, K. H., Direktor des Inst. Mineralogie, Universität, Bonn, Germany
- SCHLEITZ, N. CYRIL, Colorado School of Mines, Dept. of Metallurgical Engineering, Golden, Colorado
- SCHLECHT, WILLIAM G., U. S. Geological Survey, Washington 25, D. C.
- SCHLOCKER, J., U. S. Geological Survey, 4 Homewood Place, Menlo, California
- SCHLOSSMACHER, KARL H., Institut für Edelsteinforschung, Schliessfach 578, Idar-Oberstein 2, Germany
- SCHMIDT, ROBERT GEORGE, U. S. Geological Survey, Washington 25, D. C.
- SCHNEER, CECIL J., Geology Dept., University of New Hampshire, Durham, New Hampshire
- SCHNEIDER, PHILIP E., 46 Blue Hills Ave., Hartford 5, Connecticut
- SCHOENER, JOHN P., 22 Pollard Ave., Rochester 12, New York
- \*SCHOEP, ALFRED, 97 Baudeloo St., Ghent, Belgium
- SCHOENLAUB, R. A., 38 East Gay St., Columbus 15, Ohio
- SCHORTMANN, RAYMOND E., 306 Main Street, Easthampton, Massachusetts
- SCHULZE, JACK D., Box 555, Winslow, Arizona
- SCHWARTZ, B., 1884 Monroe Ave., Bronx 57, New York
- \*SCHWARTZ, GEORGE M., University of Minnesota, Minneapolis 14, Minnesota
- SCHWARTZ, JACK, 656 South Hendricks Ave., Los Angeles 22, California
- SCOTFORD, DAVID M., Geology Dept., Miami University, Oxford, Ohio
- SCOTT, DONALD W., Continental Sales & Equipment Co., Box 438, Hibbing, Minnesota
- SCOTT, HERBERT W., 641 Elliott Place, Glendale 2, California
- SCOTT, WILVIN E., 814 Instl. Sq., Box 248, Fairchild AFB, Washington
- SEAGER, A. F., 94 Ranelagh Rd., Ealing, London W5, England
- SEAMAN, DAVID M., Apt. B-4, 300 Richbell Rd., Mamaroneck, New York
- SEGLER, CURT G., 284 East 16th St., Brooklyn 26, New York
- \*SEIFERT, HANS, Mineralogisch-Petrographisches Institut der Westf. Landesuniversität, Münster 1 W, Germany
- SELTZER, ROBERT A., Box 222, Encino, California
- SERGIADIS, F. R., Geology Dept., De Beers Consolidated Mines, Ltd., Box 616, Kimberley, Cape Province, South Africa
- SERVILLA, THOMAS, 107 Kimball St., Iselin, New Jersey
- SERVOS, KURT M., Dept. of Geology, Yale University, New Haven, Connecticut
- \*SHAND, S. JAMES, The New Club, Princes St., Edinburgh, Scotland
- SHAPPIRIO, JOEL, 4811 17 St., N.W., Washington 11, D. C.
- SHARP, KENNETH R., Engineering Geology Section, S.M.H.E.A., Cooma, New South Wales, Australia
- \*SHAUB, BENJAMIN M., 159 Elm St., Northampton, Massachusetts
- SHAW, FRED R., Box 124, Gardnerville, Nevada
- SHELL, HASKILL R., Box 385, Norris, Tennessee
- SHERIDAN, DOUGLAS M., 2809 Xavier St., Denver 12, Colorado
- SHERMAN, WALTER C., 27 Doro Place, Rumford 16, Rhode Island
- SHIMAMOTO, K. (MRS.), 540 Trenton Ave., Cincinnati 36, Ohio
- SHIPTON, W. D., Dept. of Geology, Washington University, St. Louis 5, Missouri
- SHIROZU, HARUO, Kyushu University, Rigakubu Chishitsugaku, Kyoshitsu, Hakozakicho, Fukuoka-shi, Fukuoka-ken, Japan
- SHOEMAKER, EUGENE M., 205 Country Club Park, Grand Junction, Colorado
- SHOTT, WILLIAM L., 541 Tenth St., Oakmont, Pennsylvania
- SHURTZ, ROBERT F., 258 West Basic Rd., Henderson, Nevada
- SIDWELL, RAYMOND, Dept. of Geology, Texas Technical College, Lubbock, Texas
- SIEGRIST, MARIE, 1431 Iris St., N.W., Washington 12, D. C.

- SIEVERS, D. C., 1405 Bright Drive, Kingsport, Tennessee
- SILBERSTEIN, M. L., 1607 Francis St., Houston 4, Texas
- SILVER, LEON T., Division of Geological Sciences, California Institute of Technology, Pasadena 4, California
- SILVERMAN, JEANNE (MRS.), 1224 High St., Whittier, California
- SIMCOCK, J. H., Cranford, Lovatt Ave., Milehouse, Newcastle, Staffordshire, England
- SIMMONS, EDWARD E., JR., 455 S. Oakland Ave., Pasadena 5, California
- SIMONS, FRANK S., U. S. Geological Survey, Room 4209, GSA Bldg., Washington 25, D. C.
- SIMPSON, DAVID H., Dept. of Geology, University of Alberta, Edmonton, Alberta, Canada
- \*SINGEWALD, QUENTIN D., U. S. Geological Survey Bldg., Agricultural Research Center, Beltsville, Maryland
- SKEA, E. M., P.O. Box 46, Pilgrims Rest, Transvaal, South Africa
- SKINNER, HELEN C., Dept. of Geology, University of Adelaide, Adelaide, South Australia
- SLAVKO, MARJANOVIC, Sav. Center Za Dokumentaciju, Adm. Geprata 16, Beograd, Jugoslavia
- \*SLAWSON, CHESTER B., Mineralogical Laboratory, University of Michigan, Ann Arbor, Michigan
- SLOAN, ARTHUR M., 358 North Drive, San Antonio, Texas
- SMEDES, HARRY W., U. S. Geological Survey, So. 157 Howard St., Spokane 4, Washington
- SMITH, CHARLES H., Geological Survey of Canada, Victoria Museum, Ottawa, Canada
- SMITH, D. T., Rensselaer Polytechnic Institute, Troy, New York
- SMITH, DEANE K., JR., Geology Dept., University of Minnesota, Minneapolis 14, Minnesota
- SMITH, EDWARD S. C., Geology Laboratory, Union College, Schenectady 8, New York
- SMITH, GEORGE I., 459 N. Cambridge Ave., Claremont California
- SMITH, JOSEPH V., Dept. of Mineralogy, Downing St., Cambridge, England
- SMITH, JULIAN A., Rt. 2, Box 786B, Modesto, California
- SMITH, RAYMOND J., Dept. of Geology, Louisiana State University, Baton Rouge, Louisiana
- SMITH, ROBERT L., 4756 F. O. Blvd., Fair Oaks, California
- \*SMITH, WARD C., 550 West 12 St., Claremont, California
- SMITH, WILLIAM L., 2719 Jasper St., S.E., Washington 20, D. C.
- \*SMITH, W. S. TANGIER, 1231 Hoover St., Menlo Park, California
- \*SNELGROVE, ALFRED K., Dept. of Geology, Michigan College of Mining and Technology, Houghton, Michigan
- SNOW, ROLAND B., Room 2413, U. S. Steel Corporation, 525 Wm. Penn Place, Pittsburgh 30, Pennsylvania
- SOCLOW, ARTHUR A., Geology Dept., Boston University, 725 Commonwealth Ave., Boston, Massachusetts
- SOEDER, GUSTAVE, 28-51 42 St., Astoria 3, New York
- SOWERS, GEORGE M., Shell Oil Co., 965 First National Building, Oklahoma City, Oklahoma
- \*SPENCE, HUGH S., 222 Argyle Ave., Ottawa, Ontario, Canada
- SPENCER, CHARLES W., Box 194, Lincoln Rd., Lincoln, Massachusetts
- SPERRY, ARTHUR B., 333 Denison, Manhattan, Kansas
- SPIROFF, KIRIL, Houghton, Michigan
- SPROAT, IRA E., 58 Washington Ave., Westport, Connecticut
- STAACK, HAGEN, East Rock Rd., Allentown, Pennsylvania
- STAACK, MORTIMER H., U. S. Geological Survey, Bldg. 25, Denver Federal Center, Denver, Colorado
- STADNICHENKO, TAISIA, U. S. Geological Survey, Washington 25, D. C.



- STAHLBERG, STEN, Ymergatan 17 F, Uppsala, Sweden
- STANWOOD, CREIGHTON B., 12 Kilburn Rd., Belmont 78, Massachusetts
- \*STAPLES, LLOYD W., Dept. of Geology, University of Oregon, Eugene, Oregon
- \*STARK, JOHN T., U. S. Geological Survey, Military Geology Branch, Washington 25, D. C.
- STEAD, FRANK, Trace Elements Research Group, U. S. Geological Survey, Denver Federal Center, Denver, Colorado
- STEARNS, KENT, 7 Bachman St., Greenhills, Cincinnati 18, Ohio
- STEEN, CHARLES A., Box 402, Moab, Utah
- STEPHANO, C. S., 1016 Walnut St., Philadelphia 7, Pennsylvania
- STERN, THOMAS W., U. S. Geological Survey, Geochemistry & Petrology, Washington 25, D. C.
- STEVENS, ROBERT D., Havelock House, Northbourne Ave., Canberra City, A.C.T., Australia
- \*STEVENS, ROLLIN E., U. S. Geological Survey, Denver Federal Center, Denver, Colorado
- \*STEVENSON, JOHN S., Dept. of Geological Sciences, McGill University, Montreal, Canada
- STEVENSON, MRS. JOHN S., Redpath Museum, McGill University, Montreal, Canada
- STEWART, DAVID B., Geochemistry and Petrology Branch, U. S. Geological Survey, Washington 25, D. C.
- \*STEWART, DUNCAN, Dept. of Geology, Carleton College, Northfield, Minnesota
- STEWART, FREDERICK H., Dept. of Geology, Science Laboratories, University of Durham, Durham, England
- STEWART, GLENN W., Dept. of Geology, University of New Hampshire, Durham, New Hampshire
- STEWART, WENDELL O., 426 Stedman Place, Monrovia, California
- STIEFF, LORIN R., 9507 East Stanhope Rd., Kensington, Maryland
- STIGLER, WILLIAM J., 33 Caroline St., Lancaster, Pennsylvania
- STILWELL, STEPHEN N., 4149 Trabor Drive, Cincinnati 36, Ohio
- STIVERS, GLEN H., 350 South B St., Tustin, California
- STOBBE, HELEN, Morris House, Smith College, Northampton, Massachusetts
- \*STOIBER, RICHARD E., McKenna Rd., Norwich, Vermont
- STONE, JEROME, U. S. Geological Survey, Washington 25, D. C.
- \*STONE, ROBERT L., Dept. of Ceramic Engineering, University of Texas, Austin 12, Texas
- \*STOW, MARCELLUS H., 405 Massie St., Lexington, Virginia
- STRAW, DAVID J., Canadian Johns Manville Co., Asbestos, Quebec, Canada
- \*STRINGHAM, BRONSON F., University of Utah, Salt Lake City, Utah
- \*STROCK, LESTER W., 21 Madison Ave., Saratoga Springs, New York
- STRONG, MARTINEZ, Museo Nacional Ciencias Naturales, Paseo de la Castellano 84, Madrid, Spain
- \*STRUNZ, HUGO, Mineralogisch-Geologisches Institut, Regensburg, Germany
- STUCKER, P., Riedtalstrasse 3, Zofingen, Switzerland
- \*STUCKEY, JASPER L., 1911 Sunset Drive, Raleigh, North Carolina
- STUGARD, FREDERICK, JR., 4943 Marilyn Drive, Salt Lake City 7, Utah
- SUBRAMINIAM, A. P., Geological Survey of India, Madras 4, India
- SUDO, TOSHIO, Geol. and Min. Institute, Faculty of Science, Tokyo University of Education, Otsukakubo-machi, Bunkyo-ku, Tokyo, Japan
- SUN, MING-SHAN, Bureau of Mines and Mineral Resources, Campus Station, Socorro, New Mexico
- SUTHERLAND, J. A., 4848½ Wiota, Los Angeles 41, California
- SUTTON, WILLARD H., Mineral Industries Bldg., Pennsylvania State University, University Park, Pennsylvania
- SWINEFORD, ADA, Kansas Geological Survey, Lawrence, Kansas

SWINNEY, C. M., School of Mineral Sciences, Stanford University, California

\*SWITZER, GEORGE, U. S. National Museum, Washington 25, D. C.

\*TABER, STEPHEN, 1518 Deans Lane, Columbia 5, South Carolina

TAKAHASHI, HIROSHI, 811 Izumo-cho, Suginami, Tokyo, Japan

TAKATSUI, FUMIO, 9438 Empire Way, Seattle 8, Washington

TAKEUCHI, YOSHIO, Dept. of Physics, Pennsylvania State University, University Park, Pennsylvania

TAMURA, TSUMEO, Soils Dept., Connecticut Agricultural Experiment Station, P.O. Box 1106, New Haven 4, Connecticut

TATGE, ELEANOR, Corning Glass Works, Research Laboratory, Corning, New York

\*TAVORA, ELYSIARIO, Faculdade Nacionale de Filosofia, Universidade do Brasil, Av. Pres. Antonio Carlos 40, Rio de Janeiro, Brazil

TAYLOR, ALBERT J., 106 Placer Lane, Oak Ridge, Tennessee

TAYLOR, ALLEN O., 6286 Boxwood Rd., Salt Lake City, Utah

TAYLOR, JAMES A., 25 Old Oak Rd., Glen Ridge, New Jersey

TAYLOR, RALPH E., Box 2180, Houston 1, Texas

\*TAYLOR, W. H., The Cavendish Laboratory, Crystallographic Section, Free School Lane, Cambridge, England

TEETERS, JESSE C., Rt. 1, Springfield, Colorado

\*THIBAUT, N. W., Norton Company, Worcester, Massachusetts

THOMAS, W. HERSEY, 145 East Gorgas Lane, Mt. Airy, Philadelphia 19, Pennsylvania

THOMPSON, CLYDE L., Harbison-Walker Refractories Co., Hays Laboratory, Pittsburgh 7, Pennsylvania

THOMPSON, JAMES B., JR., Dept. of Mineralogy, Harvard University, Cambridge 38, Massachusetts

THOMPSON, MARY E., U. S. Geological Survey, Box 360, Grand Junction, Colorado

\*THOMPSON, ROBERT M., Dept. of Geology & Geography, University of British Columbia, Vancouver, B. C., Canada

THOMSON, ROBERT, Box 913, Cobalt, Ontario, Canada

THORNTON, CHARLES P., Mineral Industries Bldg., Pennsylvania State University, University Park, Pennsylvania

THURSTON, WILLIAM R., Division of Earth Sciences, 2101 Constitution Ave., Washington 25, D. C.

TILLMAN, C. G., 4 Greenough Ave., Cambridge, Massachusetts

TILTON, GEORGE R., 12608 Bluhill Road, Silver Spring, Maryland

Tischler, Martin S., U. S. Geological Survey, Box 654, Camaguey, Cuba

TITCOMB, JANE, U. S. Geological Survey, Naval Gun Factory, Washington 25, D. C.

TOLBERT, GENE E., 958 Belmont St., Watertown 72, Massachusetts

\*TOLMAN, CARL, Dept. of Geology, Washington University, St. Louis 5, Missouri

TOMLINSON, W. HAROLD, 260 North Rolling Rd., Springfield, Pennsylvania

TOOKER, EDWIN W., U. S. Geological Survey, Denver Federal Center, Denver, Colorado

TORRE ENCISO, D. EUGENIO, Plaza de Pontevedra 5, La Coruna, Spain

TOYODA, HIDEYOSHI, Matsuyama Post Office Kunai, 487, 2-chome, Dogo-Minamimachi Matsuyama, Japan

TRAINER, DAVID W., JR., 42 University Ave., Hamilton, New York

TRAUTZ, OTTO R., Dept. of Chemistry, New York University, College of Dentistry, 209 East 23d St., New York 10, New York

TRAVIS, DAVID, 5827 North Lawrence St., Philadelphia 20, Pennsylvania

TRAVIS, RUSSELL B., Colorado School of Mines, Golden, Colorado

TREFFETHEN, JOSEPH M., New Engineering Building, University of Maine, Orono, Maine

- TREMBLAY, ABBE J. A., Seminaire de Chicoutimi, Chicoutimi, P.Q., Canada
- TRICHEL, HALL, Gulf Refining Co., P.O. Drawer 1731, Shreveport, Louisiana
- TRIMBLE, CLINTON E., 308 Larkspur Ave., Corona Del Mar, California
- TRITES, ALBERT F., JR., U. S. Geological Survey, Geologic Division, Denver Federal Center, Denver, Colorado
- TROEGER, W. E., Mineralogisch Institut, Technische Hochschule, Darmstadt, Germany
- \*TRUDELL, HARRY W., 1309 Highland Ave., Abington, Pennsylvania
- TRUMPOUR, HARRY J., 1350 North 11th, Corvallis, Oregon
- TRYGGVASON, TOMAS, University Research Institute, Reykjavik, Iceland
- \*TUNELL, GEORGE, 801 Toyopa Drive, Pacific Palisades, California
- TURJA, OSCAR, 282 Birch St., Lively, Ontario, Canada
- \*TOURNEAURE, F. S., 2017 Vinewood Blvd., Ann Arbor, Michigan
- \*TURNER, FRANCIS J., Bacon Hall, University of California, Berkeley 4, California
- TURNER, MORT D., Calle 12 de Octubre, #414, Hato Rey, Puerto Rico
- \*TUTTLE, ORVILLE F., Mineral Industries Building, Pennsylvania State University, University Park, Pennsylvania
- \*TWETO, OGDEN, U. S. Geological Survey, Mineral Deposits Branch, Bldg, 25, Denver Federal Center, Denver, Colorado
- ULMER, FRED, 99 Omaha St., Dumont, New Jersey
- UYTENBOGAARDT, W., Dept., of Geology & Mineralogy, Royal Institute of Technology, Stockholm 70, Sweden
- VALENTINE, GRANT M., 1310 Rockefeller Ave., Everett, Washington
- VALENTINE, WILBUR G., Dept. of Geology, Brooklyn College, Brooklyn, New York
- VALLANCE, THOMAS G, Dept. of Geology, University of Sydney, Sydney, NSW, Australia
- VAN COTT, HARRISON C., Corning Glass Works, Corning, New York
- VAN TASSEL, HARRY C., 8009 Westmoreland Ave., Edgewood, Pittsburgh 18, Pennsylvania
- VAN DER VEEN, A. H., 13 Perziklaan, The Hague, Netherlands
- VAN HORN, EARL C., Box 221, Murphy, North Carolina
- VAN HORN, MAURICE H., Research Laboratories, Eastman Kodak Company, Kodak Park, Rochester 4, New York
- VAN VALKENBURG, ALVIN, JR., National Bureau of Standards, Washington 25, D. C.
- VAUGHAN, ALLAN C., U. S. Smelting & Refining Co., Midvale, Utah
- VENKAYYA, E., Mineralogical Institute, E.T.H., Sonneggstrasse 5, Zürich 6, Switzerland
- VERA, ELPIDIO C., Philippine Bureau of Mines, Geological Survey Division, Manila, Philippines
- \*VERHOOGEN, JOHN, Dept. of Geological Sciences, University of California, Berkeley 4, California
- VHAY, JOHN STEWART, S., 157 Howard, Spokane, Washington
- VICKERS, A. ERIC J., Wynnstay, Millfield Rd., Whickham, Newcastle-on-Tyne, England
- VIEIRA, PEDRO ALBERTINO, Depto. Nac. Producao Mineral, Seccao de Quartzzo, Avenida Pasteur 404, Rio de Janeiro, Brazil
- VILLAR FAHRE, JORGE FELIX, Paseo Colon 751, 7º Piso, Buenos Aires, Argentina
- \*VITALIANO, CHARLES J., Dept. of Geology, Indiana University, Bloomington, Indiana
- VLISIDIS, ANGELINA C., U. S. Geological Survey, Washington 25, D. C.
- VON KNORRING, OLAG, Geology Dept., The University, Leeds 2, England
- VON VULTEE, JOACHIM, Institut für Mineralogis und Lagerstättenlehre, Rhein.-Westf. Technische Hochschule, Wullnerstrasse 2, Aachen, Germany.
- VTELENSKY, JIRI, Hostovice 24, P. Zleby u. Caslavi, Czechoslovakia
- VUAGNAT, M., Onex, Geneva, Switzerland



- WADE, F. ALTON, Dept. of Geology, Texas Technological College, Lubbock, Texas
- \*WAGER, LAWRENCE R., Dept. of Geology and Mineralogy, University Museum, Oxford, England
- \*WAHLSTROM, ERNEST E., Dept. of Geology, University of Colorado, Boulder, Colorado
- WALDO, ALLEN W., 2030 North Orange Ave., Stockton 4, California
- \*WALDSCHMIDT, WILLIAM A., 1901 W. College, Midland, Texas
- WALKER, ALAN, 1040 Avenue of the Americas, New York, New York
- WALKER, FREDERICK, Dept. of Geology, University, Cape Town, South Africa
- WALL, MILFORD W., 12 Euston St., Brookline, Massachusetts
- WALLACE, ROBERT E., U. S. Geological Survey, S. 157 Howard, Spokane, Washington
- WANDKE, ALFRED D., Apartado 60, Bayamo, Oriente, Cuba
- WARRACH, HENRY C., P.O. Box 162, Hammond, Indiana
- WARSHAW, CHARLOTTE M. (Mrs.), Geology Division, Gulf Research & Development Corporation, Box 2038, Pittsburgh 30, Pennsylvania
- WASHKEN, EDWARD, 2076 Massachusetts Ave., Cambridge 40, Massachusetts
- \*WASSERSTEIN, BENNO, Geological Survey, Box 401, Pretoria, South Africa
- \*WATANABE, TAKEO, Tokyo Imperial University, Tokyo, Japan
- \*WATERS, AARON C., Geology Dept., The Johns Hopkins University, Baltimore 18, Maryland
- \*WATSON, EDWARD H., Dept. of Geology, Bryn Mawr College, Bryn Mawr, Pennsylvania
- \*WATSON, KENNETH DEP., Dept. of Geology, University of California, Los Angeles 24, California
- \*WEAYLAND, RUSSELL G., 5007 Earlston Drive, Westgate, Washington 16, D. C.
- WEAVER, CHARLES E., 3737 Bellaire Blvd., Houston 25, Texas
- \*WEBB, ROBERT W., Dept. of Physical Science, University of California, Santa Barbara, California
- \*WEEKS, ALICE M. (Mrs.), Geochemistry & Petrology Branch, U. S. Geological Survey, Washington 25, D. C.
- WEEKS, CHARLES A., Red Gate Lane, Meredith, New Hampshire
- WEEKS, WILFORD F. 2/LT., AO 2237725, Hq. USAF Cambridge Research Center, L. G. Hanscom Field, Bedford, Massachusetts
- WEHRENBURG, JOHN P., Dept. of Geology, Montana State University, Missoula, Montana
- WEIDHAAS, ERNEST, 255 Corlies Ave., Pelham 65, New York
- WEIS, PAUL L., U. S. Geological Survey, 157 South Howard St., Spokane, Washington
- WEISS, E. JOSEPH, Dept. of Ceramic Engineering, University of Texas, Austin 12, Texas
- WEITZ, JOHN H., Independent Explosive Co., 455 Leader Bldg., Cleveland 14, Ohio
- WELLNITZ, ANNA M., 90 Elwood Place, Staten Island 1, New York
- WELLS, RALPH G., U. S. Steel Corporation, Applied Research Laboratory, Monroeville, Pennsylvania
- WELSH, DOROTHY, 3003 P St., N.W., Apt. 302, Washington 7, D. C.
- WELT, RUTH M., 132 Bay 31st St., Brooklyn 14, New York
- WENDEN, HENRY E., 16 Normandy Rd., Woodhaven, Lexington 73, Massachusetts
- \*WEST, CUTLER D., Polaroid Corporation, 718 Main St., Cambridge 39, Massachusetts
- \*WHERRY, EDGAR W., 137 Maplewood Ave., Philadelphia 44, Pennsylvania
- WHITAKER, GEORGE C., 825 East Schaaf Rd., RD 3, Brooklyn Station, Cleveland, Ohio
- WHITE, DAVID A., Bureau of Mineral Resources, Turner Hostel, Canberra City, Australia
- WHITE, J. F., 132 Riddle Road, Cincinnati 15, Ohio
- WHITE, W. ARTHUR, Illinois State Geological Survey, Urbana, Illinois
- WHITE, W. E., 552 Booth St., Ottawa, Canada
- \*WICKMAN, FRANS E., Stockholm 50, Sweden

- WILK, BIRGER, Fredsgatan 11-C-42, Helsinki, Finland
- WILCOX, RAY E., Geologic Division, Denver Federal Center, Denver, Colorado
- WILD, GEORGE O., Idar Oberstein 2, Germany
- WILKERSON, ALBERT S., Dept. of Geology, Rutgers University, New Brunswick, New Jersey
- WILLEMS, H. W., 1513 Pacific Ave., Breckenridge, Pennsylvania
- WILLIAMS, EDWARD E., 20463 Riopelle, Detroit 3, Michigan
- WILLIS, WILLETT R., 1343 North Nevada Ave., Colorado Springs, Colorado
- WILLOW, STEPHEN B., 2 West 47th St., New York 36, New York
- WILLMARTH, VERL R., U. S. Geological Survey, Mineral Deposits Branch, Washington 25, D. C.
- \*WILSON, ARTHUR J. C., University College, Cardiff, South Wales, Great Britain
- WILSON, FORD E., 2924 Mountain Ave., El Paso, Texas
- \*WINCHELL, ALEXANDER N., 88 Vineyard Rd., Hamden 17, Connecticut
- \*WINCHELL, HORACE, 40 Deepwood Drive, Hamden 17, Connecticut
- \*WINKLER, HELMUT G. F., Mineralogisches Institut der Universität, Deutschhausstr. 10, Marburg/Lahn (16), Germany
- WINNER, MAURICE D., JR., 2352 Gruver Ave., Easton, Pennsylvania
- WINTRINGHAM, NEIL A., 1485 Force Drive, Mountainside, New Jersey
- WISE, WILLIAM S., School of Mineral Sciences, Stanford University, Stanford, California
- WISHART, JAMES S., 56 Hammond St., Rochester 13, New York
- WITTELS, MARK C., 108 Amherst Lane, Oak Ridge, Tennessee
- WOLBERS, JOSEPH W., 585 West 16th St., San Pedro, California
- \*WOLFE, C. WROE, Dept. of Geology, Boston University, Boston, Massachusetts
- WOLKODOFF, VLADIMIR E., 4215 Lamar St., Wheat Ridge, Colorado
- WOO, C. C., Crane Co., 836 South Michigan Ave., Chicago, Illinois
- \*WOOD, ELIZABETH ARMSTRONG (MRS.), Bell Telephone Laboratories, Murray Hill, New Jersey
- WOOD, LEONARD E., Dept. of Geology, Michigan State University, East Lansing, Michigan
- \*WOODFORD, ALFRED O., 443 West 10th St., Claremont, California
- WOODHOUSE, C. D., 4125 Creciente Drive, Santa Barbara, California
- WOODS, ARTHUR E., Route 1, Box 182, Clearwater, Florida
- WOODTLI, ROBERT, B. P. 18, Nizi, Ituri, Belgian Congo
- WOODWARD, THEODORE, 4207 Hewitt Ave., Silver Spring, Maryland
- WOOLARD, LOUIS E., New Jersey Zinc Co., Grand Junction, Colorado
- WOYSKI, MARGARET SKILLMAN (MRS.), Lindsay Light & Chemical Co., West Chicago, Illinois
- WRIGHT, F. W., JR., Orinoco Mining Co., Apartado 3, Ciudad Bolivar, Venezuela
- WRIGHT, HAROLD D., 711 West Hamilton Ave., State College, Pennsylvania
- WYART, J., Laboratoire de Mineralogie, Faculté des Sciences, 1 Rue Victor Cousin, Paris, France
- WYATT, MICHAEL, School of Mineral Sciences, Stanford University, Stanford, California
- WYMAN, RICHARD V., Box 27, Leeds, Utah
- YAGI, KENZO, Institute of Earth Sciences, First College of Arts and Sciences, Tohoku University, Tomizawa, Sendai, Japan
- \*YAGODA, HERMAN, Industrial Hygiene Research Laboratory, National Institute of Health, Bethesda 14, Maryland



- YAMAGUCHI, MASAMICHI, Nagoya Univ. Rigakuhu, Chikyu-Kagaku-Kyushitsu, Furocho, Chikusa-ku, Nagoya, Japan
- YAMAGUCHI, MASARU, Dept. of Geology, Faculty of Science, Kyushu University, Fukuoka, Japan
- YAMAMOTO, HIROSATO, Kurume Branch, Fukuoka-Cakugei University, 105 Nishimabhi, Kurume City, Fukuoka Pref., Japan
- YAMAMOTO, TAKASHI, Geology Dept., Kyushu Institute of Technology, Tobata City, Fukuoka Pref., Kyushu, Japan
- YASHIMA, RYUICHI, c/o Chigaku-Kenkyushitsu, Fukushima-Daigaku-Gakugeigakubu, Hamadacho, Fukushima, Japan
- YEDLIN, LEO N., 129 Englewood Drive, New Haven, Connecticut
- \*YODER, HATTEN S., JR., Geophysical Laboratory, 2801 Upton St., N. W., Washington 8, D. C.
- YOKOYAMA, KANAE, c/o Torimi, 12 Tamayashita, Sendai, Japan
- YORKS, KENNETH P., 104 Ormsby Drive, Syracuse 4, New York
- YOST, CHESTER H., Mio, Michigan
- YOUMANS, K. A., Box 575, Sitka, Alaska
- YOUNG, HOWARD S., R.D. 1, Chadds Ford, Pennsylvania
- YOUNG, JOHN A., JR., Sun Oil Co., Production Dept., 1608 Walnut St., Philadelphia 3, Pennsylvania
- YOUNG, ROBERT SPENCER, Virginia Geological Survey, Box 1428, University Station, Charlottesville, Virginia
- YOUNG, W. FORD, P.O. Box 187, Belize, British Honduras
- \*ZACHARIASEN, WILLIAM HOULDER, Dept. of Physics, University of Chicago, Chicago 37, Illinois
- ZAGORKA, STOJANOVIC, Zav. Za Geol. Rud. Tehn. Istrazavanja, Nemanjina ul. 6, Beograd, Yugoslavia
- ZAK, LUBOR, Mineralogical Dept., Charles University, Prague 2, Czechoslovakia
- ZANNARAS, J. P., Box 500, Congress, Arizona
- ZEIHEN, LESTER G., 834 West Silver St., Butte, Montana
- ZEMANN, JOSEF, Nonnenstieg 22, Göttingen, Germany
- ZEN, E-AN, 42 Kirkland St., Cambridge 38, Massachusetts
- \*ZERFOSS, SAMUEL, National Bureau of Standards, Mineral Products Division, Washington 25, D. C.
- ZIMMER, PAUL W., Irontown, Minnesota
- ZIMMERLE, WINFRIED, Richmond Petroleum Company of Colombia, Apartado Nac 2760, Bogota, Colombia
- ZIMMERMAN, RICHARD A., 75 Athol St., Springfield, Massachusetts
- \*ZODAC, PETER, Box 28, Peekskill, New York
- ZUNINO, CARLOS, Calle Rivadavia 1173-1175, San Juan, Argentina